December 2019

Status and remediation of mercury in fish and aquatic ecosystems

Geoffrey Dean Millard
Syracuse University

I. Abstract

Mercury is a known neurotoxin which can bioaccumulate and biomagnify as methylmercury, causing significant human and ecological impacts. New York State and the surrounding region is impacted by mercury in complex ways as some areas are recovering from the effects of acidic deposition while the entire region is increasingly influenced by climate change. In order to better understand the drivers and trends of mercury dynamics in New York State, my research was conducted in three phases: 1) the impact of lime (calcium carbonate) additions on mercury cycling in small acid-sensitive headwater streams of the Adirondack State Park; 2) applied advanced analytical techniques to improve our understanding of the complex relationship between mercury and dissolved organic matter; and 3) examined spatial and temporal trends of fish mercury concentrations across New York State

Given interest in the relationship between acidification and fish mercury concentrations, in phase one I examined the use of lime as an acidification remediation strategy and its impact on mercury cycling. Lime has been shown to effectively mitigate the effects of acidic deposition in northern Europe as well as northeastern North America, however the impact of this management strategy on mercury cycling has not been examined. Previous work at Honnedaga Lake has shown a watershed scale application of lime alters mercury transport and cycling and is highly correlated with dissolved organic matter in headwater streams. Extending the period of record demonstrates that elevated mercury and dissolved organic matter in streamwater draining a limed watershed continued for the three years of my study. This pattern contrasts with streams that received direct channel applications which resulted in much more limited increases lasting only 72 - 96 hours. In these small headwater streams, mercury was mobilized from treated areas but was not methylated before being exported from the study site. Critically, there was no evidence of increased bioaccumulation in stream macroinvertebrates following treatment, however my

study period was too short to examine the long-term impacts and potential community changes resulting from continued calcium amendments.

In phase two, I explored the relationship between mercury and dissolved organic carbon (DOC). Following the watershed addition at Honnedaga Lake, a shift in the specific ultraviolet absorbance of DOC was observed. This shift suggests an increased molecular weight and aromaticity of DOC entering the limed tributary. Several studies have shown that the quality of dissolved organic matter can have important effects on the bioavailability of mercury and methylmercury. Using advanced analytical techniques and methods developed from laboratory experiments, I analyzed aquatic samples collected from the watershed treatment and reference streams. Preliminary analysis of surface waters by Fourier Transform Ion Cyclotron Resonance Mass Spectroscopy (FTICR-MS) reveals an increase in the proportion of thiol functional groups and aromaticity in the first four weeks following treatment, but no significant difference between study sites in later samples. This information could help inform watershed management decisions and serve as a proxy for natural recovery of aquatic ecosystems from acidic deposition.

Phase three built upon a previous state-wide survey of lake fish conducted from 2003-2005. This survey found significant biological mercury hotspots in the Adirondack and Catskill State parks, and significant correlations of fish mercury concentrations with factors related to acidification and wetland area. The latest survey, conducted from 2013-2015, I found somewhat different results. Fish Hg in the Adirondack and Catskill regions remained elevated relative to the rest of NYS, however, the landscape and chemistry drivers from the earlier survey no longer applied across the whole state. Long-term changes in fish mercury concentrations were not evident in the recent survey. This lack of a change may suggest the major driver of fish mercury for inland waters is shifting away from regional Hg emissions towards the effects of legacy Hg and acid inputs or changes in nutrient status, invasive species, climate change and/or increasing global Hg emissions.

Status and remediation of mercury in fish and aquatic ecosystems

by

Geoffrey Dean Millard

B.S., St. Lawrence University, 2010
M.S., Syracuse University, 2016

Dissertation

Submitted in partial fulfillment of the requirements for the degree of

Doctor of Philosophy in Civil Engineering.

Syracuse University

December 2019

II. Acknowledgements

Financial support for the research contained within this dissertation was in part provided by the New York State Energy Research and Development Authority, the National Science Foundation Research Trainee program and the United States Department of Energy. This work could not have been completed without collaborations with researchers at the United States Geological Survey, the New York State Department of Environmental Conservation Cornell University, SUNY Environmental Science and Forestry, SUNY Oneonta, Oak Ridge National Lab and the Environmental Molecular Sciences Laboratory at the Pacific Northwest National Lab.

I would like to thank my advisor, Dr. Charles Driscoll, for his guidance throughout this process and for providing me with many opportunities to advance my professional goals. I also appreciate the guidance and support of my committee members: Dr. Svetoslava Todorova, Dr. Teng Zeng, Dr. Baohua Gu, Dr. Andria Costello-Staniec, Dr. Laura Lautz and Dr. Chris Junium. Thank you to Mario Montesdeoca for his constant excitement and encouragement and my fellow graduate students, Steve Boucher, Habib Fakhraei, and Caitlin Eger for their friendship over the past six years. I would also thank Mariah Taylor and Amy Shaw for the countless hours they spent processing and analyzing fish samples.

Finally, I would like to thank my family. My parents, Jane and Pete Millard always encouraged and supported my academic pursuits and my two brothers Matthew and Michael who not only acted as sounding boards, but tirelessly keep my ego in check. Most of all, I would like to thank my wife Stacey and my sons Oliver and Isaac for their constant love and support. They faced the challenges of pursuing a Ph.D. with me and never wavered in their belief of what I could achieve.

III. Table of Contents

IV. List of Figures

V. List of Tables

1. Introduction

Mercury (Hg) is a naturally occurring element in the Earth's biogeochemical system. Human perturbations such as mining and fossil fuel consumption have dramatically increased mercury (Hg) levels in the biosphere (Selin 2009). Recent estimates suggest 2320 Mg yr^{-1} of Hg are emitted to the atmosphere by human activities (Pirrone et al. 2010, UN Environment 2019; Figure 1). The current rate of anthropogenic and legacy re-emissions is 2-15 times greater than that from natural geogenic sources (Driscoll et al. 2013) Once in the atmosphere, Hg can distribute globally by deposition followed by rapid re-emission, making it a widespread and persistent pollutant.

Areas with considerable canopy cover adsorb and absorb Hg through the stomata of leaves, resulting in high levels of Hg in litterfall and throughfall (Hintelmann et al. 2002, Blackwell and Driscoll 2015, Gerson et al. 2017) before being mobilized into aquatic ecosystems (Skyllberg et al. 2003). Mercury bound up with dissolved organic matter can be metabolized by sulfate or iron reducing bacteria or archaea producing methylmercury (MeHg) (Gilmour et al. 1998, Kerin et al. 2006, Dittman 2010). This organic form of mercury strongly bioaccumulates and biomagnifies in ecosystems (Benoit et al. 2002).

In addition to increased anthropogenic emissions, mercury cycling has also been affected by anthropogenic forcing of climate and historical acidic deposition. Climate change impacts have been observed in the northeastern United States with increased air temperature, precipitation and streamflow (McCabe and Wolock 2002, Hodgkins et al. 2003, Huntington et al. 2004, Climate Science Special Report 2017). These changes could result in elevated Hg methylation due to increased lake productivity (Watson et al. 2016), causing eutrophication and longer anoxic periods.

Also in the northeastern United States, surface water acidification by historical inputs of acidic deposition have caused elevated bioaccumulation of MeHg (Yu et al. 2011). Acidic deposition in the form of sulfate (SO_4^{2-}) can drive the formation of MeHg by sulfate-reducing bacteria (Drevnick et al. 2007, Coleman Wasik et al. 2015). The success of regulations reducing acidic emissions have resulted in increased concentrations of DOC (Monteith et al. 2007) and in some cases resulting in a coincident increase in Hg concentrations in fish (Hongve et al. 2012).

Despite decreases in U.S. Hg emissions (Drevnick et al. 2012, Zhang et al. 2016) and some declines in Hg concentrations and deposition in precipitation (Prestbo and Gay 2009, Mao et al. 2017, Zhou et al. 2018), fish Hg concentrations in remote ecosystems have generally remained elevated (Driscoll et al. 2007b, Simonin et al. 2008a, French et al. 2014). Some ecosystems where fish Hg previously responded to reductions in U.S. emissions are reversing trends in response to the increasing importance of climate and global emissions (Zhou et al. 2017). It is critical that we understand the drivers and mechanisms of mercury transport and bioaccumulation in order to mitigate and manage the environmental and human health risks.

In order to improve our understanding of mercury dynamics, I have organized this dissertation into three phases. In phase one, I evaluate and compare the impact of watershed and channel $CaCO_3$ additions on Hg stream transport, production of MeHg, and MeHg accumulation in aquatic macroinvertebrates at Honnedaga Lake watershed. Although calcium additions have been studied extensively in Europe (Appelberg and Degerman 1991, Bradley and Ormerod 2002, Clair and Hindar 2015) and North America (Newton et al. 1996, Fiorentino et al. 2003, Cho et al. 2009, Lawrence et al. 2016); changes in Hg dynamics resulting from Ca addition remain relatively unknown. I hypothesize that 1) compared to in-stream additions, the watershed addition would have a longer-lasting impact on Hg transport and would mobilize a larger mass of

Hg from the watershed to the tributary and the lake; and 2) after the first in-stream addition, the subsequent in-stream additions would mobilize decreasing amounts of Hg as hydrologically connected stocks of labile DOC become depleted. I examined MeHg in macroinvertebrates because the food webs of these streams are dominated by macroinvertebrates (most are fishless), and because macroinvertebrate MeHg concentrations have been strongly correlated with concentrations of dissolved MeHg in other Adirondack streams (Riva-Murray et al. 2011). Thus, an increase in MeHg in stream water as a result of liming could result in an increase in concentrations of MeHg in macroinvertebrates of treated streams.

In phase two, I examined two small first order tributaries to Honnedaga Lake. Both of these streams were chronically acidic with pH below 5 until treatment with $CaCO_3$. Following a watershed-scale application, Millard et al. (2018) reported maximum DOC concentrations in the treated stream of 18.4 mg/L during the first post-treatment year. In addition to elevated DOC concentrations, a significant shift in specific ultraviolet absorbance (SUVA) was observed following the lime addition. This pattern suggests not only a change in the quantity of DOC, but also the quality.

Several recent studies have applied Electrospray Ionization Fourier Transform Ion Cyclotron Resonance-Mass Spectrometry (ESI-FTICR-MS) to characterize the molecular features of organic sulfur, despite a low abundance in the DOM pool (Sleighter et al. 2014, Pohlabeln and Dittmar 2015, Gomez-Saez et al. 2016, Ksionzek et al. 2016). ESI-FTICR-MS has ultrahigh mass resolving power and mass accuracy and has become a powerful technique for the molecular characterization of complex mixtures like DOM (Koch et al. 2005, Sleighter and Hatcher 2007, Hertkorn et al. 2008). I hypothesize that the $CaCO_3$ treatment will cause an increase in DOM with thiol functional groups. This elevated DOM would have a higher molecular weight and

increased aromatic character than pre-treatment and reference samples. These changes in DOM will result in elevated transport of Hg from soils to the stream.

Phase three of my dissertation places the work of the first two phases in a broader context and builds upon a previous New York State survey of lake fish conducted in the mid-2000s. This earlier survey found significant biological mercury hotspots in the Adirondack and Catskill State parks, and significant correlations of fish mercury concentrations with factors related to acidification (pH, conductivity, ANC), water-Hg concentrations, wetland area and the presence of dams. In this phase, I evaluate changes in NYS fish Hg concentrations over the last decade by resurveying 43 lakes that were sampled in the earlier study. Fish were also collected from 65 previously unsampled lakes. Using information from both surveys, recommendations were developed for a long-term monitoring program to assess the efficacy of Hg emission reduction efforts in New York State. These recommendations include locations where fish Hg and ancillary observations should be monitored, the number of waters monitored and frequency of collections. I hypothesize there will be a general decline in fish Hg across the state, with the largest improvements in the Adirondack and Catskill regions. However, the Adirondack and Catskill regions will still have widespread Hg contamination compared to other regions of the state due to enhanced deposition associated with forest cover and acidic deposition recovery. Furthermore, surface waters with relatively large riparian and wetland areas will have higher fish mercury concentrations.

2. Literature Review

2.1 Mercury Emissions

As a naturally occurring element, Hg is released to the atmosphere through both natural and anthropogenic processes. Natural sources, such as volcanic activity and mineral weathering account for a relatively small percentage of new emissions because human perturbations such as mining and fossil fuel consumption have dramatically increased mercury levels in the biosphere (Selin 2009). Recent estimates suggest that Hg loads to the atmosphere have increased by 450% because of human activity, predominantly coal combustion and artisanal gold mining. These increases are thought to be driven by a better accounting for emissions, a plateau in reductions in North America and Europe, and the continuing industrialization of developing regions (Streets et al. 2018, UN Environment Programme Chemicals and Health Branch 2019).

The current rate of anthropogenic and legacy re-emissions is 2-15 times greater than from natural geogenic sources (Driscoll et al. 2013) and have continued to increase by approximately 20% between 2010 and 2015 (UN Environment Programme Chemicals and Health Branch 2019). Once in the atmosphere, elemental Hg can distribute globally by depositing and rapidly re-emitting, making it a widespread and persistent pollutant. Estimates of atmospheric residence time for elemental Hg have been adjusted downward by about 10% to 0.93 years because of better understanding of elemental Hg oxidation by atmospheric bromine (Seigneur and Lohman 2008). Emissions of ionic and particulate mercury tend to deposit regionally due to residence times of days to weeks, but these species can be converted to elemental mercury and re-emit to the atmosphere (Selin 2009, Driscoll et al. 2013).

2.2 Mercury Deposition and Transport

Mercury is transported widely by atmospheric processes and distributed through terrestrial and aquatic ecosystems (Driscoll et al. 2013). Concentrations of Hg in precipitation and wet Hg deposition have been declining in the eastern U.S. over the recent decade in response to reductions in North American emissions (Prestbo and Gay 2009, Mao et al. 2017). However it has recently been demonstrated that global emissions and long-range transport are of increasing importance to North American deposition (Weiss-Penzias et al. 2016, Streets et al. 2019). In the northeastern United States, an estimated 60-80% of Hg deposition has been attributed to regional sources of gaseous or particulate ionic Hg (Driscoll et al. 2007b, Selin 2009). Forested areas, in particular, act as atmospheric sinks with considerable canopy surface area for Hg adsorption and absorption through the stomata of leaves. This foliar enrichment of Hg facilitates elevated deposition to soils via litterfall and throughfall (Hintelmann et al. 2002, Blackwell and Driscoll 2015, Gerson et al. 2017). Soils retain Hg bound to reduced sulfur groups associated with organic matter until the organic matter is mobilized in water (Haitzer et al. 2002, Skyllberg et al. 2003).

Mercury transport through the aquatic environment occurs largely in association with dissolved or particulate organic matter (Dittman et al. 2010). In reducing environments, Hg can be converted to methylmercury (MeHg) by sulfate- or iron-reducing bacteria or archaea (Gilmour et al. 1998, Kerin et al. 2006). This organic form of Hg strongly bioaccumulates and biomagnifies along aquatic and terrestrial food chains (Benoit et al. 2002). The relationship of dissolved organic matter, often referred to as dissolved organic carbon (DOC), and MeHg is complex. Natural DOC has been shown to facilitate the formation of MeHg (Graham et al. 2012), but also reduce bioaccumulation rates (Gorski et al. 2008).

2.3 Mercury toxicity

The bioaccumulation and biomagnification of Hg remains a major concern. Methylmercury is readily taken up at the base of aquatic food webs. Concentrations magnify along food chains resulting in elevated concentrations that can pose neurotoxicological risks to humans, mammals and fish- and insect-eating birds (Driscoll et al. 2007b, 2013). As a potent neurotoxin, the consequences of human and environmental exposure are internationally recognized with collective efforts to reduce exposure via the Minimata Convention (Minimata Convention on Mercury 2017). The major pathway of human exposure is through the consumption of contaminated fish (Driscoll et al. 2013). Approximately 95% of MeHg consumed is absorbed through the gastrointestinal tract (Aberg et al. 1969). After entering the bloodstream, MeHg can cross the blood-brain and blood-fetus barriers, causing neurological damage in humans and other top predators (Burgess and Meyer 2008, Syversen and Kaur 2012, Schoch et al. 2014). It has been estimated the global economic cost from only health impacts associated with Hg will reach 3.7 billion USD by 2020 (Sundseth et al. 2010) with the total economic impact to the U.S. could be 4.8 billion USD per year (Grandjean and Bellanger 2017), while the economic benefit of reducing Hg emissions could reach 104 billion USD by 2050 in the U.S. alone (Giang and Selin 2016). Another study found that reducing methylmercury exposure in the U.S. by as much as 10% could realize an annual economic benefit of 860 million USD (Rice et al. 2010)

2.4 Fish Mercury in New York State

The bioaccumulation and biomagnification of Hg is a major concern across New York State (NYS; Dittman et al. 2010). With over 4,000 lakes in NYS, measurement of fish Hg concentrations is important to track environmental, economic and health impacts. A survey in

the mid-2000s of 131 lakes divided NYS into three broad regions (Southeast, Northeast and West) found 53 lakes with fish Hg concentrations high enough for the NYS Department of Health to issue consumption advisories while considering U.S. FDA and U.S. EPA guidelines (1µg/g and 0.3µg/g, respectively; Simonin et al. 2008b). Fish Hg concentrations were negatively correlated with pH, conductivity, and acid neutralizing capacity (ANC), and positively correlated with water-Hg concentrations, wetland area, and the presence of dams. Spatially, the Adirondack and Catskill regions of NYS were found to have the highest fish Hg.

In the Northeastern U.S., concentrations of DOC have been increasing, possibly in response to recovery from historical acidic deposition (Monteith et al. 2007, Driscoll et al. 2016). This pattern is particularly important in regions like the Adirondack Park where decades of SO_4^{2-} deposition have depleted available calcium (Ca) and other base cations from soils (Warby et al. 2005), delaying recovery from acidification (Driscoll et al. 2001, 2003, 2007a; Chen and Driscoll 2005). A lengthy recovery could result in an extended period of contamination as fish Hg concentrations have been shown to increase with decreases in pH and increases in DOC (Driscoll et al. 2007b).

While recovery from acidic deposition appears to be an important factor contributing to increases in fish Hg, anthropogenic forcing of climate can also affect bioaccumulation (Evans et al. 2006, Sebestyen et al. 2009). In the northeastern U.S., climate change is expected to generate higher annual temperatures, earlier peak streamflow from snowmelt, a longer growing season, increased drought frequency and increased winter precipitation (Hayhoe et al. 2007a, Climate Science Special Report 2017). Effects of climate change have been observed in the Northeastern U.S. with increases in air temperature, precipitation and streamflow (McCabe and Wolock 2002, Hodgkins et al. 2003, Huntington et al. 2004, Climate Science Special Report 2017). These

changes could increase lake productivity (Watson et al. 2016), causing eutrophication and extending lake stratification, which would promote Hg methylation due to increased temperature or an increased anoxic period in lake sediments. An amplified hydrological cycle could also increase wet-dry cycles stimulating methylation rates (Coleman Wasik et al. 2015). While the long term impacts of climate change remains unclear (Laudon et al. 2012), there has been some potential evidence of increased fish Hg (Bodaly et al. 1993, Monson et al. 2011, Zhou et al. 2017).

2.5 Acidic Deposition and Remediation

In the northeastern U.S., streams are showing ANC recoveries of <1 μeq L^{-1} y^{-1} (Burns et al. 2006, Lawrence et al. 2011, Likens and Buso 2012). In areas that have started to recover from acid deposition, researchers have reported an increase in DOC coinciding with increases in fish Hg concentrations (Hongve et al. 2012). Despite some recovery, episodic acidification continues to occur during high flows (Fuss et al. 2015). Episodic acidification impacts are pronounced in acid-sensitive areas like the Adirondack region of New York, where models predict ecosystem recovery will take multiple decades (Waller et al. 2012, Fakhraei et al. 2014). In a 2003-2005 study of western Adirondack streams, over half of stream reaches experienced episodic acidification that was sufficiently intense to mobilize toxic concentrations of dissolved inorganic aluminum (Lawrence et al. 2008). Long-term inputs of acidic deposition have resulted in depletion of available base cations from forest soils, therefore addition of basic materials to soils and surface waters have been explored as an approach to accelerate ecosystem recovery from acidic deposition. Previous studies have shown that Ca additions in the form of wollastonite ($CaSiO_3$) and agricultural lime ($CaCO_3$) can accelerate the recovery of acid-impacted watersheds (Cirmo and Driscoll 1996, Driscoll et al. 1996, Cho et al. 2009, Shao et al. 2016). Calcium base

additions modify soil and aquatic chemistry by increasing exchangeable Ca in soils resulting in increases in pH and ANC, and decreased concentrations of inorganic monomeric aluminum (Lawrence et al. 2016).

Direct addition of $CaCO_3$ to streams has been shown to substantially improve water quality over the short-term (Clair and Hindar 2005). Reproduction and survivability of Brook Trout (*Salvelinus fontinalis*) have recovered following this type of addition, but other species of fish and macroinvertebrate communities have not replicated this success (Hall et al. 1994, Menendez et al. 1996, Clayton et al. 1998, Hudy et al. 2000). Despite repeated $CaCO_3$ additions, treated streams could be falling short of full recovery due to fluctuations in water chemistry over the length of the channel, periods of high flowrate, or the limited duration of direct stream additions (Lawrence et al. 2016). Watershed-scale $CaCO_3$ and other types of Ca additions have been used to improve soil and water quality over longer periods. These treatments have been shown to increase the base status of soils for a decade or more (Dalziel et al. 1994, Bradley and Ormerod 2002, Hindar et al. 2003, Johnson et al. 2014). This enhancement in soil quality can mitigate acidification effects on forest vegetation (Battles et al. 2014) and reduce concentrations of aluminum and other metals in soil and stream waters, further improving aquatic conditions (Grieve 1990, Fransman and Nihlgaard 1995). These watershed-scale treatments have been shown to decrease fish mercury concentrations in downstream lakes (Håkanson et al. 1988, Rask et al. 2007, Shastri and Diwekar 2008).

2.6 Natural Organic Matter Complexes with Mercury

Organic carbon, or more broadly natural organic matter (NOM) is an important ligand for the transport of Hg in natural systems in both dissolved and particulate forms (Dittman et al. 2010), with one group showing that sulfur-containing functional groups could dominate NOM

binding of Hg (Chen et al. 2017). Studies have shown NOM can increase (Graham et al. 2012) or decrease (Gorski et al. 2008) the bioavailability of Hg under different conditions. Under anoxic conditions, Graham et al. (2012) found high concentrations of NOM result in higher MeHg production from SO_4^{2-} reducing bacteria *in vitro*. They also found that MeHg was strongly associated with NOM following production. In natural systems, Gorski et al. (2008) found lower bioconcentration factors when NOM was greater than 5 mg/L and suggest that high NOM inhibits bioavailability, while low concentrations aid algal uptake of MeHg.

This seemingly inconsistent role of NOM suggests that Hg bioavailability can be mediated by a variety of factors including: redox conditions, NOM concentration, and bacterial species. Existing ubiquitously in aquatic environments, NOM is chemically heterogeneous and redox reactive (Aiken et al. 1985, Nurmi and Tratnyek 2002, Chen et al. 2002, Cory and McKnight 2005). Natural organic matter containing thiol-functional groups strongly bind with ionic Hg (Skyllberg et al. 2000, 2006; Dong et al. 2011) and are the dominant ligand in natural aquatic ecosystems (Chen et al. 2017). Pure culture studies of sulfate-reducing bacteria under sulfidic conditions significantly enhanced methylation over a range of Hg:DOM ratios (Graham et al. 2012, 2013; Moreau et al. 2015) and suggests that high molecular weight aromatic DOM limits the formation of HgS nanoparticles, potentially enhancing bioavailability (Graham et al. 2012, 2013). This pattern contrasts with a recent study in a series of lakes in southwest China where lower molecular weight autochthonous DOM resulted in higher bioaccumulation factors (Jiang et al. 2018).

A study in the Adirondacks hinted at the possibility of high DOM inhibiting fish mercury concentrations, but lacked data from a range of high DOM lakes (Driscoll et al. 1994). More recently, French et al. (2014) examined water and amphipod mercury concentrations in 26 lakes

in the Canadian arctic ranging from <3mg-C L^{-1} to over 18 mg-C L^{-1}. This study revealed two interesting findings. First, the bioconcentration factors increased until DOC concentrations of ~8.5mg-C L^{-1} and then decreased at higher concentrations. This impact level is close to the value suggested by Gorski et al. (2008). Second, the fraction of total Hg bound to larger aromatic acids, dramatically increased at ~11.1 mg-C L^{-1}. While there are not many lakes in the Adirondacks with reported DOC concentrations greater than this threshold (Dittman and Driscoll 2009, Roy et al. 2012), Driscoll et al. (1994) reported one lake with DOC concentrations of ~28 mg/L and very low fish mercury concentrations.

3. Phase 1: Liming comparison at Honnedaga Lake tributaries

3.1 Methods

3.1.1 Study Site

Honnedaga Lake (3.1 km^2 surface area), located in the southwestern Adirondacks (43º31'06"N and 74º48'31"W) was highly impacted by acidic deposition which nearly extirpated a genetically-unique "heritage" Brook Trout population. With decreases in acidic deposition, conditions in the lake have improved and the population has re-colonized the lake (Josephson et al. 2014). However, spawning and nursery areas are still limited due to chronic and episodic acidification of tributaries. To enhance recruitment of a more robust population, $CaCO_3$ was applied to the watershed of a chronically-acidic tributary in October, 2013, and to the stream channels of two episodically-acidic tributaries annually from 2012 to 2016. This treatment was expected to reduce acidity and improve water quality, while providing an opportunity to examine effects of watershed and in-stream lime applications on mercury mobilization and bioaccumulation.

The 13.3 km^2 Honnedaga Lake watershed is completely forested, with 26 tributaries that drain into the lake. The six tributaries examined in this study (i.e., three limed and three reference; Figure 2) ranged from episodically to chronically-acidic (pH<5.0) prior to treatment (Josephson et al. 2014). Two episodically-acidic tributaries received direct stream application (EL1, EL2) and one chronically-acidic tributary received a watershed scale application (CL1) of $CaCO_3$. Two episodically-, and one chronically-acidic tributary were used as reference tributaries (ER1, ER2, CR1)

The limed watershed (CL1) received 150 metric tons of limestone (mainly $CaCO_3$), distributed by helicopter in a pelletized form over the 30-hectare watershed at a dose of 1.4 Mg

of Ca/ha during early October, 2013 as part of a larger multi-disciplinary ecosystem study (Homan et al. 2016; Millard et al. 2018; George et al. 2018). This dosing level was comparable to application rates for other regional watershed liming studies (Driscoll et al. 1996; Peters et al. 2004). Streams that experienced direct channel additions received annual treatments of 11T at EL1 (12 July 2012, 19 June 2013, 28 Feb 2014, 16 June 2015, and 21 June 2016) and 5T at EL2 (12 July 2012, 19 June 2013, 28 Feb 2014, and 16 June 2015) based on "West Virginia" and "Clayton" formulas (Schmidt and Sharpe 2002). Before the onset of this study, EL1 was treated with 5.5T of lime on 28 Oct 2010. As a result, EL1 received two additional treatments than EL2, including the second in 2016. These five tributaries represent 22% of total lake watershed area (Total – 8.72 km^2 CL1 – 0.19 km^2, CR1 – 0.11 km^2, EL1 – 1.35 km^2, EL2 – 0.19 km^2, ER2 – 0.11 km^2). The second episodically-acidic reference tributary (ER1 – 1.69 km^2) is not within the Honnedaga Lake watershed, but directly adjacent and with similar watershed characteristics to EL1.

3.1.2 Field Sampling of Streams

Water samples were collected manually for analysis of major ions, pH, and DOC at approximately two-week intervals. Grab samples were supplemented using automated water samplers beginning in January 2012. These samplers were programmed to collect water samples that represented the rising, peak, and recession hydrograph during storm events and snowmelt. Field blanks and triplicate samples were collected for additional quality control. Water samples for Hg analysis were collected seasonally beginning in the spring of 2012, when sample locations were physically accessible. Following lime addition at CL1, Hg samples were collected weekly at CL1 and CR1 during the first four weeks, and approximately monthly thereafter through September 2016. Sample collection for Hg analysis in episodically-acidic

tributaries was monthly, except for two brief periods in 2015 and 2016 at EL1, when samples were collected multiple times a day and at ER2, where Hg samples were only collected three times in 2012 and once in 2014. Stream samples for Hg were collected using trace-metal clean techniques (U.S. EPA method 1669, U.S. EPA 1995). Simultaneously with each Hg sample, a separate water sample was also collected for analysis of DOC, SO_4^{2-}, nitrate (NO_3^-), iron (Fe) and pH. After treatment, two new Hg sample sites were added, EL1R and EL2R, located above the $CaCO_3$ addition locations on EL1 and EL2, respectively. A new site, EL1M, was also added just below the $CaCO_3$ addition point on EL1. In 2016, an additional sample location, EL1P, was added on EL1 approximately 10 m upstream of another wetland complex and downstream of the $CaCO_3$ addition point (Figure 2; Table 1). Water samples were transported on wet ice to a field laboratory, where the Hg samples were passed through 0.4 µm polycarbonate filters and acidified to 1% with concentrated hydrochloric acid prior to shipping to the Syracuse University (SU) laboratory. Samples for analysis of DOC, ultraviolet absorbance at a wavelength of 254 nm (UV_{254}), SO_4^{-2}, NO_3^-, Fe and pH were transported on ice to the USGS New York Water Science Center Laboratory in Troy, NY (NYWSC) and processed for analysis according to procedures described at: https://www.sciencebase.gov/catalog/item/55ca2fd6e4b08400b1fdb88f (accessed 3/14/2019).

Macroinvertebrate samples were collected during mid-May (spring) in 2012 and 2014, May 31 – mid-June (early summer) in 2013-16, and July – early August (mid-summer) every year. Targeted macroinvertebrates were crayfish (Cambaridae), and larvae of aquatic insects, particularly dragonflies (Odonata), northern caddisflies (Trichoptera: Limnephilidae), filtering caddisflies (Trichoptera: Hydropsychidae), and shredding stoneflies (Plecoptera: Leuctridae and Nemouridae); other taxa were collected as well (Table 2). Specimens were collected from all

available habitats, with nets and by hand-picking, using trace metal clean techniques. Specimens were placed in site water in acid-cleaned plastic jars or new plastic bags, kept cool, and were processed at a field laboratory (generally within several hours of collection; always < 24 hours). Field processing included sorting by taxon and (in some cases) by size into composite samples, rinsing ≥ three times in deionized water, placing into new tared plastic containers, weighing, and freezing; samples were kept frozen until laboratory processing. Additional details are provided elsewhere (Scudder et al. 2008, Riva-Murray et al. 2011).

3.1.3 Laboratory Processing and Chemical Analyses

Water samples were analyzed at the USGS Mercury Research Laboratory (MRL; samples collected during 2012) or the Water Quality Laboratory (all subsequent samples) at SU. Water samples were sent to MRL for filtered THg and MeHg. Filtered THg was determined using cold vapor atomic fluorescence spectrometry (CVFAS) according to the method described by Olson and DeWild (1999), which is a slight modification to EPA method 1631 (U.S. EPA 2002). Filtered MeHg was determined after distillation by ethylation, gas chromatographic separation, pyrolysis and CVAFS as described in DeWild et al. (2002); this is a modification of EPA method 1630 (U.S. EPA 2007a).

Water samples were analyzed post-2012 for filtered THg and MeHg at SU. Total Hg was analyzed using an oxidation, purge and trap, desorption and CVFAS following U.S. EPA method 1631, revision E (U.S. EPA 2002); the method detection limit is 0.2 ng/L. Methylmercury was analyzed by distillation, ethylation, purge and trap, desorption and CVFAS according to U.S. EPA Method 1630 (U.S. EPA 2007a); the method detection limit is 0.02 ng/L. For both THg and MeHg in water continuing calibration verification (CCV) standards and blanks were analyzed every ten samples with additional matrix spikes (MS, MSD) and duplicates analyzed

every twenty samples. An additional ongoing precision recovery (OPR) samples were analyzed at the beginning and end of each batch (Appendix 1).

Water samples were analyzed at the NYWSC for pH, DOC, UV_{254}, SO_4^{2-}, NO_3^-, and Fe. Dissolved SO_4^{2-} and NO_3^- were analyzed by ion chromatography after first passing the subsample through a 0.4 µm polycarbonate filter. UV_{254} and DOC were analyzed by UV-enhanced persulfate oxidation in a total carbon analyzer, after first passing the subsample through a 0.7 µm glass fiber filter (GFF). UV_{254} was determined using a standard spectrophotometer (Weishaar et al. 2003) and corrected for Fe interference, as described by Poulin et al. (2014). Specific ultraviolet absorbance (SUVA) was calculated from measured absorbance at 254 nm per unit DOC concentration. Dissolved Fe was analyzed by Inductively Coupled Plasma (ICP) spectrophotometry, after first passing the subsample through a 0.4 µm polycarbonate filter and then acidified with ultrapure nitric acid. An unfiltered subsample was also analyzed for pH in the laboratory. Further NYWSC method details and QA/QC procedures can be found at https://www.usgs.gov/centers/ny-water/science/laboratory (accessed 03/14/2019).

3.1.4 *Determination of Stream Solute Loads*

Stream stage was measured by the USGS at each site every 15 minutes with a submersible pressure transducer. Stage was converted to discharge through the use of a stage-discharge rating curve developed by making periodic discharge measurements with a current meter under a range of flow conditions. Methods for measuring stream stage and discharge are described in Sauer and Turnipseed (2010), and Turnipseed and Sauer (2010).

Annual loads of SO_4^{2-}, DOC, THg and MeHg were determined using scripts developed by W. David Watkins and Alison Appling with funding from the USGS (‘*LoadflexBatch*’

https://github.com/USGS-R/loadflexBatch). LoadflexBatch builds off of previously developed USGS tools '*rloadest*' (Runkel and De Cicco 2017) to estimate loads and '*loadflex*' (Appling et al. 2015) to reduce bias. These packages are derived from the LOADEST statistical modeling platform and are commonly used to evaluate loads based on continuous discharge and intermittent chemistry data. *'LoadflexBatch'* calculates an annual (water-year) load for each chemical parameter of interest over multiple sites and years within a record. The RL7 model provided the best statistical fit and has the general form:

$$\ln(C) = 1 + T + T^2 + \ln(Q) + \ln(Q)^2 + \sin(T) + \cos(T)$$

where C = concentration, T = decimal time, and Q = discharge. This concentration – discharge regression was applied to calculate an estimated mean concentration for each day, which when multiplied by mean daily discharge yielded a daily load value. These daily loads were then used to estimate an annual load ± standard error for each tributary. Discharge from the tributaries over the study period is shown in Figure 3.

3.1.5 Analysis of Hg and Nutrient Concentration Data

Water samples were divided into groups based on treatment period. At CL1, pre-treatment observations were assigned to period 1. Time series of THg, MeHg and DOC concentrations were used to identify transitional- (Oct 2013-Feb 2014) and post-treatment (March 2014 – Oct 2016) response periods (2 and 3, respectively) after lime addition in CL1. Stream-limed water samples were divided into periods between each addition of $CaCO_3$. Pre-treatment samples were assigned to period 1 with new periods beginning after each $CaCO_3$ addition. The EL1 tributary had been limed two years before my study by personnel at Cornell University and as such no true pre-treatment observations are available. For this site, stream measurements begin at period 1* and end with period 6. The EL2 tributary had not been

previously treated and so begins with period 1 and ends with period 5, as it was not treated in 2016 (Table 3). Samples were paired either by grab sampling in all the tributaries or by storm events from the USGS automated samplers. Some of these samples were below detection limits when analyzed and were excluded from statistical tests. Paired samples of THg, MeHg, DOC, SO_4^{2-} and UV_{254} were collected at each tributary.

Statistical analyses of water data were performed with the software package Rv3.4.2 (R Core Team 2018). Regression analyses were performed on concentration data from each stream to describe THg and MeHg relationships with DOC, SO_4^{2-}, UV_{254} and specific ultraviolet absorbance (SUVA; UV_{254}/DOC). A standard ANOVA was applied to all pre-treatment observations at CL1 and CR1, including unpaired data, to examine statistical differences in the mean concentrations of SO_4^{2-}, NO_3^-, DOC, UV_{254}, SUVA, THg, MeHg and %MeHg (MeHg*100/THg) among these streams. The unpaired data were included to compare MeHg pre-treatment values because paired data were not available for this analyte. Standard ANOVA was used to test pre-treatment DOC and SO_4^{2-} loads among sites. Using the aov function from the statistical package in R, a least square means (LSM) fixed model

$$Y = Site + Period + Site * Period$$

was used to test the main effect of site and treatment and examine for interaction. In this model Y is a response metric (THg, MeHg, DOC, etc.), site is sample location (CL1 , EL1, ER1, etc.) and period is the time interval between lime additions for episodically-acidic streams, and the previously defined intervals for chronically-acidic streams (Millard et al. 2018). Where the LSM model indicates probable interaction between site and period (α=0.2), a before-after control-impact (BACI) paired design was used with an ANOVA test in the function *glht* from the R package *multcomp* v1.4-8, to assess changes in the difference between sites over the treatment

periods. A p-value of less than or equal to 0.05 was considered significant and less than 0.1 marginally-significant.

3.1.6 Macroinvertebrate Hg and Stable Isotope Analyses

Most MeHg analyses of macroinvertebrate samples were conducted at SU; a small number of samples from 2012 were analyzed at MRL. Samples analyzed at SU were freeze-dried, ground, and analyzed by distillation, ethylation, purge and trap, desorption and CVFAS according to U.S. EPA Method 1630 (U.S. EPA 2007a) after being extracted in 25% potassium hydroxide at 60^{o}C for 48 hours. Every 20 samples were extracted with a blank, duplicate, matrix spike, matrix spike duplicate and duplicates of National Institute of Standards and Technology Standard Reference Material 2976 (mussel tissue) and Canadian National Research Council Standard Reference Material Tort-2 (lobster tissue). Extracts were analyzed with continuous calibration verification every ten samples (Appendix 2). Macroinvertebrate samples analyzed for MeHg at MRL were freeze-dried, ground, and analyzed by cold-vapor atomic fluorescence spectroscopy after extraction by dilute nitric acid following Hammerschmidt and Fitzgerald (2006).

After analysis for MeHg, the remaining freeze-dried and ground sample material was submitted for $\delta^{15}N$ and $\delta^{13}C$ isotope analyses. Samples collected during 2012 and 2013, were analyzed at the Environmental Science Stable Isotope Laboratory (EaSSIL) at the State University of New York College of Environmental Science and Forestry (Syracuse, NY). EaSSIL used a Costech elemental analyzer coupled to a Thermo Finnigan Delta XP-Plus isotope ratio mass spectrometer. Samples collected after 2013 were analyzed by the Cornell University Stable Isotope Laboratory (COIL; Ithaca, NY) using a ThermoFinnigan Delta Plus mass spectrometer. Accuracy and precision of the stable isotope measurements at both laboratories

(expressed in the standard per mil notation relative to Vienna Pee Dee Belemnite [V-PDB] for $\delta^{13}C$ and atmospheric nitrogen for $\delta^{15}N$) were verified by reference materials provided by the International Atomic Energy Association (IAEA). Daily precision of the instrument was verified by repeated analysis of internal laboratory standards during the sample runs. Additional QA/QC information can be found in the Table 4.

3.1.7 Analysis of Macroinvertebrate Hg and Stable Isotope Data

Macroinvertebrate sample results were analyzed to compare the pattern of change in MeHg concentrations, normalized to a common trophic position of 2.5 (henceforth "$MeHg_{2.5INV}$") among treated and untreated sites, and before and after $CaCO_3$ treatments. Trophic-position (TP) normalization was done using the following formulae (Post 2002, Anderson and Cabana 2007):

$$MeHg_{2.5INV} = \left(\frac{MeHg}{TP_{base-adjusted}}\right) * 2.5$$

$$TP_{base-adjusted} = \frac{\left(\delta^{15}N_{sample} - \delta^{15}N_{base}\right) + 1}{3.4}$$

The base consumer was selected after inspecting dual isotope bivariate plots ($\delta^{15}N$ x $\delta^{13}C$) for each site (data in Table 2); $\delta^{15}N_{base}$ was the mean of all samples. Base consumers were northern caddisflies (Trichoptera: Limnephilidae; 8 sites), filtering caddisflies (Trichoptera: Hydropsychidae; 1 site), and swimming mayflies (Ephemeroptera: Isonychia; 1 site).

Comparisons were conducted with a combination of graphical analysis (boxplots) and group comparison tests using exact Wilcoxon (for two-group comparisons) or analysis of variance on ranks followed by Tukey's test (for comparisons of $\geq$ 3 groups). Statistical tests were not conducted on groups containing fewer than five samples. Northern caddisflies were not included in comparison of early-summer (i.e., pre-liming) $MeHg_{2.5INV}$ with late-summer (i.e.,

post-liming) $MeHg_{2.5INV}$ because they were rare in the latter collections. Significant difference for the macroinvertebrate analyses was defined as $p < 0.05$, and p-values ≥ 0.05 and < 0.1 were considered marginally-significant.

3.2 Results

3.2.1 THg and MeHg Relations with Water Quality Parameters

Over the entire study period, THg concentrations were significantly positively related with DOC concentrations at CL1, EL1, EL1M, EL1R, EL2, CR1, and ER1 (adj-R^2 = 0.83, 0.263, 0.1519, 0.1918, 0.4644, 0.63, 0.2556 respectively; Table 5). Positive regressions between THg and UV254 were also significant at the same sites CL1, EL1, EL1M, EL1R, EL2, CR1, and ER1 (adj-R^2 = 08453, 0.3834, 0.1031, 0.1685, 0.483, 0.5194, 0.1651). Significant positive relationships between THg and SUVA occurred only at EL1 (adj-R2 = 0.1504) and CL1 (adj-R2 = 0.3751). Organic monomeric Al was significantly positively correlated with THg at all the treatment sites (CL1, EL1, and EL2; adj-R^2 = 0.1711, 0.4236, and 0.3294) and ER1 (adj-R^2 = 0.3467), while pH and specific conductance were significantly positively related with THg at CL1 only (adj-R^2 = 0.2055, 0.2556, respectively).

Concentrations of MeHg were significantly positively related to THg only at EL1 and EL1R (adj-R^2 = 0.5898, 0.297). MeHg was significantly positively correlated with DOC and UV254 at EL1 (adj-R^2 = 0.2171, 0.2713) and EL1P (adj-R^2 = 0.3322, 0.3934) and was significantly positively correlated with SUVA at EL1M and EL1P (adj-R^2 = 0.3472, 0.2616). Additional information can be found in Table 5.

3.2.2 Watershed $CaCO_3$ Treatment (CL1 and CR1)

Watershed lime addition resulted in immediate increases in concentrations of THg, DOC, SUVA, SO_4^{2-} and NO_3^- following treatment and remained significantly elevated through the first

water year compared to reference values as previously reported (Millard et al. 2018). After the watershed application, pH values peaked at 7.46 immediately after application at CL1 and then declined continually to episodically acidic values of ~5.0 by May 2016 (32 months after application). These pH values, however remained elevated compared with the reference stream (CR1; Figure 4). A gradual decrease in the treatment response of NO_3^- concentrations was also evident in a comparison of NO_3^- at CL1 and CR1, until no difference was evident between these streams by 2014 (summary statistics Table 6).

Despite declining pH and NO_3^- concentrations in the treated watershed after the first year, DOC, UV254, SUVA, and SO_4^{2-} have remained significantly elevated relative to the reference watershed through the end of 2016 ($p<0.01$). Following treatment, THg concentrations also remained significantly higher at CL1 ($p<0.001$), with the THg:DOC ratio somewhat higher (marginally-significant; $p=0.055$). This pattern aligns with the more distinct post-treatment DOC, SUVA, and UV254 patterns ($p<0.001$, 0.008, and <0.001, respectively), which indicates the enhanced mobilization of DOM at the treated watershed (CL1) relative to the reference watershed (CR1) and pre-treatment observations, coincides with increases in THg. The highest observed DOC concentration was 18.6 mg/L and occurred in October 2016 at CL1.

The temporal pattern of MeHg concentrations indicated a short-term increase during the first five months after treatment (Millard et al. 2018). Following this period, ending in May 2014 no significant LSM interaction between CL1 and CR1 was detected for the duration of the study period. However, both tributaries experienced a major increase in MeHg concentrations and percent MeHg in 2016, when observations reached new maximum values (CL1 – 0.26 ng/L, 21%; CR1 – 0.17 ng/L, 14%, Figure 5).

Post-treatment seasonal loads of SO_4^{2-}, NO_3^- and DOC did not differ significantly between CL1 and CR2. THg loads in the winter, summer and fall of 2014, and again in the winter of 2016 were significantly higher at CL1 than at CR1. MeHg loads were significantly higher at CL1 in the fall 2013 through the spring of 2014. After this initial difference, MeHg loads were not significantly different between CL1 and CR1 (Figure 6). No significant differences were found for water years 2014 through 2016.

3.2.3 Direct Stream $CaCO_3$ Treatment (EL1, EL2, ER1, ER2)

In-stream lime additions at EL1 resulted in a significant LSM interaction for pH compared with EL1R ($p<0.001$) during the study period, with maximum values occurring during period 2, immediately after application (Figure 4). LSM interactions between EL1 and EL1R were also significant for THg ($p=0.099$), DOC ($p=0.05$), UV254 ($p=0.003$) and SUVA ($p<0.001$). BACI tests found that pH was significantly higher in the limed streams during each treatment period following period 2 ($p<0.004$). BACI tests also revealed no change in THg:DOC following the first treatment, but significantly higher THg:DOC ratio in period 3 than in all the periods that followed ($p<0.05$). Concentrations of THg declined marginally from period 2 to period 4 ($p=0.071$) and remained significantly lower in period 5 ($p=0.038$) and marginally lower in period 6 ($p=0.054$), but not in period 7 ($p=0.311$). Marginally-significant differences in THg occurred among periods 3-5 ($p=0.061$), 3-6 ($p=0.086$), and 5-7 ($p=0.081$; Table 7). The final treatment period showed a marginally-significant increase in THg over the previous two periods ($p=0.081, 0.121$).

Results of BACI tests for THg and UV_{254} did not have a similar pattern to the DOC results except that period 7 was significantly higher than all previous periods ($p<0.015$); there were no differences in DOC among other periods. UV254 did not significantly change from

period 1 to period 2 ($p>0.2$), but declined significantly from period 3 until period 5 and then increased (Figure 7). Similarly, SUVA gradually declined until the fifth period, before increasing again (Figure 8). In summary, pH saw a significant sustained increase, while THg, UV254, and SUVA saw gradual decreases over time until period 7 when these parameters along with DOC increased.

Many of these changes were also detected for differences between EL1 and EL1M. Significant LSM interactions were detected for THg, %MeHg, THg:DOC, pH, DOC, UV254, and SUVA ($p<0.2$). This LSM interaction resulted in only marginal differences in THg; in period 3 THg was marginally greater than in periods 5 and 6 ($p= 0.064, 0.086$ respectively) and in period 5 was marginally lower than period 7 ($p=0.077$). Similarly, %MeHg was significantly lower in period 5 than in period 6 ($p=0.018$) and marginally lower than in period 7 ($p=0.079$). For Hg:DOC, the difference between sites in period 3 was significantly greater than 4, 5, and 6 ($p<0.05$) and marginally greater than 7 ($p=0.053$).

The difference in pH was significantly greater in period 5 than 3, 4, and 7 ($p<0.05$), and significantly greater in period 6 than 4 ($p=0.007$). The difference in DOC for period 7 was significantly greater than in period 3 ($p=0.009$) and marginally greater than 5 and 6 ($p=0.071$, $p=0.052$, respectively). The pattern of UV_{254} was similar to DOC, with the difference in period 7 significantly greater than periods 4, 5, and 6 ($p<0.01$) and marginally greater than period 3 ($p=0.072$). BACI tests split SUVA into two groups with period 3 and 7 significantly greater than periods 4, 5 and 6 ($p<0.05$). In summary, there were gradual increases in THg and MeHg during the last three treatment periods, while %MeHg continued to decline. DOC, UV_{254} and SUVA all saw increases in period 7 relative to earlier periods.

Unlike the broad differences observed in water chemistry patterns between EL1 and EL1R, there were only limited differences between EL1 with ER1. Significant LSM interactions were only detected for DOC and UV254 ($p<0.1$). These LSM interactions were driven by period 7 which had a marginally/significantly greater difference than periods 4, 5, and 6 (DOC: $p= 0.085, 0.022, 0.006$ respectively; UV254: $p<0.05$). UV_{254} also had a marginally-significant difference between period 3 and 6 ($p=0.086$).

At EL2 and EL2R, the only significant LSM interactions were found for MeHg ($p=0.198$), pH ($p<0.001$), and SUVA ($p=0.005$). For MeHg, period 3 was significantly greater than 4 ($p=0.020$) and marginally greater than 1 and 5 ($p<0.08$). The difference in pH significantly increased, while SUVA significantly decreased from period 1 observations. These shifts were maintained for the duration of the study ($p<=0.006$, Figure 4) and no significant difference was detected between periods 2, 3, 4 and 5 (pH: $p>0.1$, SUVA: $p>0.9$).

Statistical differences between EL2 and ER2 revealed significant LSM interaction for pH ($p<0.001$) and THg ($p=0.145$) and THg:DOC ($p=0.041$). BACI contrasts showed that all the treatment periods had significantly higher pH than period 1 ($p<0.001$), with period 3 having significantly higher pH than periods 2 ($p=0.045$) and period 5 ($p=0.010$). The BACI contrasts only indicate period 4 was significantly higher than both periods 1 ($p=0.003$) and 2 ($p=0.050$), while Hg:DOC in period 1 was marginally lower than 2 ($p=0.079$) and significantly lower than 4 ($p=0.002$). (Other comparisons could not be made because THg samples were only collected at ER2 during periods 1, 2 and 4).

Seasonal and annual loads at all these sites showed no significant differences through time ($\alpha=0.05$). However, graphical analyses indicate less seasonal variability in MeHg, THg, DOC and SO_4^{2-} loads at EL1 during 2016 than other years (Figure 5, 9-11). In addition, THg

and MeHg loads appear to increase somewhat at EL2 during the fall of 2016 (although this was not statistically different from other seasonal estimates).

3.2.4 EL1 2016 Intensive sampling period

The intensive stream sampling following the 2016 EL1 channel addition revealed sharp, rapid increases in concentrations of DOC, THg, and MeHg (Figure 12). During this pulse, maximum values were observed approximately 36 hours after calcium carbonate addition (DOC = 16.8 mg/L; THg = 3.24 ng/L; MeHg = 2.59 ng/L). These increases were relatively short-lived, with concentrations returning to pre-application levels within 72 to 96 hours (DOC = 11.1mg/L; THg = 1.80 ng/L; MeHg = 0.31 ng/L).

3.2.5 Macroinvertebrate Methylmercury ($MeHg_{2.5INV}$) Response Patterns

Trophic-position-normalized MeHg in macroinvertebrates ($MeHg_{2.5INV}$) from CL1 (i.e., watershed-limed site) did not differ significantly between pre-liming (i.e., 2013) and post-liming (2014-16) collections, in early summer (Figure 13A) or late summer (Figure 13B). Similar pre-liming and post-liming $MeHg_{2.5INV}$ was observed in early summer at the five untreated sites (Figure 13A), and in mid-summer at three of the five untreated sites. Significant annual variation in mid-summer $MeHg_{2.5INV}$ occurred only at EL1R and ER1, the two open-canopied sites ($F=2.7$ and $p = 0.04$, $F = 3.1$ and $p = 0.04$, respectively), but pre-liming (i.e., 2013) $MeHg_{2.5INV}$ was significantly higher than post-liming $MeHg_{2.5INV}$ only at ER1 (Figure 13B).

Five sites had enough samples for graphical and (or) statistical comparison of early summer (prior to stream liming) $MeHg_{2.5INV}$ with mid-summer (about a month after June liming) $MeHg_{2.5INV}$ in 2013, 2015, and 2016, as well as in 2014 (when $CaCO_3$ was applied in February to episodic streams). Neither reference site exhibited a significant difference between seasons in any year except for 2013, when $MeHg_{2.5INV}$ appeared graphically to be somewhat higher in mid-

summer at ER1 (Figure 14A) and was significantly higher in mid-summer at EL1R (Figure 14B, $p = 0.02$). The treated site closest to the stream lime addition location (EL1M) had significantly higher $MeHg_{2.5INV}$ in mid-summer collections than in early-summer collections during 2013 ($p = 0.0004$) and 2016 ($p = 0.014$), but $MeHg_{2.5INV}$ did not differ significantly between collection periods in 2014, when there was no intervening liming (Figure 14C). The next site further downstream of the application point, but above the large wetland complex (EL1P, only sampled in 2016), also had significantly higher post-liming $MeHg_{2.5INV}$ in 2016 (Figure 14D, $p = 0.014$). In contrast, the downstream-most site (EL1, located downstream of the wetland) exhibited only a marginally-significant increase in $MeHg_{2.5INV}$ in mid-summer during 2016 (Figure 11 E, $p = 0.051$).

Pairwise comparisons of $MeHg_{2.5INV}$ in 2014 with other treatment years, within seasons, revealed no significant differences between 2014 and any other year in early summer at any stream liming site, whether reference or treatment (Figure 15). However, significantly higher mid-summer $MeHg_{2.5INV}$ occurred at EL1M in treatment years other than 2014 (2013, $p = 0.0006$; 2016, $p = 0.009$; insufficient samples to test 2015). In addition, somewhat higher $MeHg_{2.5INV}$ occurred in 2016 than in 2014 at ER1 (marginally-significant, $p = 0.051$), although there was no significant difference in mid-summer $MeHg_{2.5INV}$ between 2014 and either 2013 or 2015 at this site. The large difference observed between 2014 and 2013 at EL1M in mid-summer contrasts with the pattern of no significant difference between 2014 and other years at either reference site.

3.3 Discussion

3.3.1 Chemical response to watershed and stream liming

Elevated THg concentrations at the watershed limed site (CL1) reflects leaching associated with DOM from upland soils as a result of decreases in partitioning of DOM to soil (Andersson et al. 1999, Ussiri and Johnson 2004, Dittman et al. 2010) caused by increased soil pH. DOC and THg released from upland soils was transported to the stream without sustained methylation. Generally, methylation occurs in riparian and hyporheic zones within watersheds (Skyllberg et al. 2003, Tsui et al. 2008), but these zones are limited in the CL1 watershed. Increases in SUVA and the absence of a bioaccumulation response (i.e., $MeHg_{2.5INV}$) suggest that mobilized THg entering CL1 is associated with less labile allochthonous organic matter, making it less bioavailable for methylation within the stream channel or the landscape.

This interpretation is supported by the chemical response during the first water year after watershed liming which showed significant increases in THg, DOC, UV_{254} SUVA, SO_4^{2-}, and NO_3^- (Millard et al. 2018). There was a short period of five months when MeHg concentrations were elevated in CL1 relative to CR1, suggesting a limited capacity for mobilization of MeHg, but the rapid return to reference concentrations indicates that this capacity was rapidly exhausted in this upland stream setting (Millard et al. 2018). Extending the period of record at CL1 and CR1 revealed that the chemical changes observed in the first water year continued to remain elevated for the entire study period, with the exception of NO_3^-, which eventually returned to pre-treatment levels. Nitrate has been shown to limit methylation in lacustrine systems (Matthews et al. 2013), suggesting that there may be broader downstream impacts to watershed scale $CaCO_3$ additions as SO_4^{2-} and THg remain elevated.

The chemical response from direct stream additions was different and more varied than that of the watershed application. The watershed treatment resulted in longer-term increases in DOC, SO_4^{2-}, NO_3^- and THg relative to the channel additions. A spike in DOC and THg was evident immediately following each channel addition. In contrast to the sustained elevated concentrations at the watershed-treated site (CL1), the in-channel treatment sites experienced a pulse in THg concentrations of markedly shorter duration (i.e., 72-96 hours; Figure 13) in 2016, but the less-intensive sampling schedule in other years precluded the identification of this short-duration pulse. The decline in THg, UV254 and SUVA at EL1 following each direct application likely indicates a fairly rapid depletion of available and readily leachable dissolved organic matter and its associated Hg from hydrologically connected pools, resulting in shorter and less intense responses to each subsequent stream addition.

EL1 drains the largest watershed among the streams that were treated in this study, and has the greatest percentage of wetland area. One wetland is located approximately 20 m upstream of the lime addition point and another intervening wetland located just downstream of the second intermediate treatment site (EL1P). This relatively large wetland complex coupled with short hydrologic transport distance could explain the inability to detect any LSM interaction with MeHg, as wetlands are important sources of MeHg to streams and were not directly treated in this study (Dennis et al. 2005, Simonin et al. 2008a, Graham et al. 2012, Burns et al. 2012). This upland wetland influence could also explain some of the interannual variability observed in THg:DOC and %MeHg at EL1. The lack of biotic response at the site furthest downstream of the treatment location also could be due in part to the dominant influence of the intervening wetlands.

The pattern at EL1 contrasts with EL2, which does not have the same wetland influence. The relatively high dissolved silicon concentrations at EL2 (average = 3.23 mg Si/L; approximately 2x other streams) suggest this stream is influenced by groundwater sources. This stream only responded to the winter application (Feb 2014), when MeHg concentrations significantly increased and the timing of peak concentrations shifted to earlier in the season (Figure 5). This stream also experienced a significant decrease in SUVA following the initial lime application which was sustained for the entire study period and possibly indicates an increase in algal productivity.

These differences in response suggest that watershed application of lime can increase solute loads to downstream lakes and rivers relative to channel applications. The elevated SO_4^{2-} and THg could increase methylation in the downstream lake, if DOC is not sufficiently stable and elevated to decrease the bioavailability of ionic Hg (Gorski et al. 2008, French et al. 2014, Herrero Ortega et al. 2017, Mangal et al. 2019). Channel applications resulted in a pulse response, and a more rapid return to pre-liming conditions that required repeated applications to maintain desired pH levels. Additionally, the source of streamwater (surface vs. ground water) seems to play a critical role in the chemical response to direct stream applications.

3.3.2 Impact of watershed and stream liming on Hg bioaccumulation

The similarity in macroinvertebrate $MeHg_{INV2.5}$ within seasons at the watershed-limed site (CL1) before and after treatment indicates the absence of any sustained impact of watershed liming on Hg bioaccumulation over the duration of the study. This pattern corresponds with the very short duration (< 5 months, only until late winter) of the observed increase in aqueous MeHg at CL1 after watershed liming. A change in bioaccumulation could still occur over a longer period as a result of other environmental changes associated with watershed liming,

particularly decreases in acidity. An increase in pH could produce long-term changes in the food web base (e.g., more primary production), longer food chains, and/or more biodiversity and broader food webs. If this were to occur, the direction and extent of change in MeHg bioaccumulation would depend on the form of food web changes occurring over time. Greater bioaccumulation could result from longer food chains (Riva-Murray et al. 2011) and/or a shift away from mainly terrestrial carbon (i.e., leaf litter) to greater use of periphyton as an energy source (Riva-Murray et al. 2013). Conversely, less bioaccumulation could result from increased macroinvertebrate biomass or density (Chen and Folt 2005a), greatly increased periphyton biomass (Hill and Larsen 2005), and (or) faster macroinvertebrate growth rates (i.e., growth dilution; Karimi et al. 2010). Nonetheless, I did not observe any of these potential long-term changes in MeHg bioaccumulation during this study.

Some macroinvertebrate community changes have been reported at this site after liming, particularly a decline in biomass in the year following watershed liming (likely the result of physical disruption by the application), followed by a continual recovery through 2016 (George et al. 2018). A decline in macroinvertebrate biomass might be expected to result in an increase in MeHg bioaccumulation due to the concentration of available MeHg in a smaller quantity of macroinvertebrate biomass, but this was not observed. This could be due to the timing of the short-term increased MeHg concentrations, or that there is little in-stream productivity in this system.

The densely-canopied forested setting of the watershed-limed site could have contributed to the lack of bioaccumulation response over the duration of the study. The only yearly variation in $MeHg_{2.5INV}$ bioaccumulation observed occurred during mid-summer in the open-canopied reference sites that were noted (personal observation) to have greater macroinvertebrate

diversity, longer food chains, and more periphyton growth than the heavily-shaded (and more acidic) sites. Thus, there is more opportunity later in the growing season for annual variation in factors such as DOC and temperature to impact MeHg availability and bioaccumulation than earlier in the growing season. In addition to the short duration of increased aqueous MeHg at the watershed-limed site, bioconcentration at the base of the food web could have been limited by dense shading and limited primary production. Patterns of MeHg bioaccumulation could be different after watershed liming in streams with greater light exposure driving more in-stream productivity.

Comparing seasonal and annual (i.e., within season) patterns in macroinvertebrate $MeHg_{2.5INV}$ in the stream-limed tributary sites with those at reference locations hints at a possible impact of liming. However, this effect appears to be limited, both temporally and spatially. Evidence for an increase in MeHg bioaccumulation associated with liming is strongest at the site immediately downstream of the lime application (i.e., EL1M); which had significantly higher macroinvertebrate $MeHg_{2.5INV}$ about a month post-liming than in pre-treatment collections during summer-treated years. Notably, this seasonal difference was not observed in 2014, when the treatment was applied in February. Additional evidence of a treatment effect after liming occurred in 2016 at the next site downstream (EL1P), which also had significantly higher macroinvertebrate MeHg after summer liming than in the pre-liming collection that year. If these results are, indeed, an indication of a bioaccumulation response to lime application, it is most likely a result of the observed short-lived increase in aqueous MeHg concentrations after in-stream treatments. The lack of any pattern of increasing $MeHg_{2.5INV}$ over time within seasons as liming on an annual basis continued from 2013 to 2016 indicates that any effect on

bioaccumulation of this temporally-limited increase in bioavailable MeHg is also likely to be short-lived in this system.

Any effect of the in-stream liming, via an increase in bioavailable MeHg, appears to be spatially-limited as well in this system, corresponding with the spatially-limited increase in aqueous MeHg observed (i.e., mainly at sites immediately downstream of the addition, but upstream of the large wetland). A spatially-limited response to the treatment is indicated by the occurrence of higher post-liming macroinvertebrate MeHg at the two sites below treatment (i.e., EL1M, EL1P), and the lack of a significant increase at the site located far downstream and below the wetland (i.e. EL1). Overall, these results indicate that addition of lime to these relatively small, densely-forested Adirondack streams, whether by watershed treatment or annual in-channel additions, has, at best, temporally- and spatially- limited effects (increases) on MeHg bioaccumulation during the study period. A longer period of observation (and in-channel liming), during which the macroinvertebrate and periphyton communities could show change associated with improvements in water quality, would be needed to assess any potential food-web mediated effects of liming on MeHg bioaccumulation.

3.3.3 Broader Implications

Watershed and direct stream applications of $CaCO_3$ have significantly different impacts on aquatic Hg dynamics. One important difference is the increase in the concentrations and transport of DOC in stream drainage. Both applications caused significant changes in THg concentrations, however only the watershed treatment caused a sustained significant increase in DOC and THg concentrations. This increase in surface water DOC and THg may be due to a decrease in the partitioning of DOC due to increases in soil pH (Ussiri and Johnson 2004). Although there are limited observations to suggest a response of surface water Hg, widespread

observations indicate increases in DOC following recovery from elevated acidic deposition (Monteith et al. 2007, Hongve et al. 2012, Driscoll et al. 2016), which suggests the possibility of enhanced mobilization of DOC-associated THg transport in forested watersheds (Dittman et al. 2010). As natural recovery continues in regions previously impacted by acidic deposition, there will likely be complimentary effects from climate change (Clark et al. 2010), potentially coinciding with enhanced mobilization of DOM and THg.

Climate change has and is predicted to increase air and surface water temperature, the frequency of drought, and the quantity and intensity of precipitation (Hayhoe et al. 2007b, Climate Science Special Report 2017). These changes could further increase the loading of DOC to streams (Sebestyen et al. 2009) and associated Hg. Increases in temperature and precipitation will likely alter methylation rates as well. For example, an intensified hydrology cycle could increase the frequency of rewetting events that have been shown to drive methylation in wetlands (Coleman Wasik et al. 2015). This change in wet-dry cycles may explain the sudden increase in MeHg across most of our study sites in 2016 (Figure 5). In 2015 and 2016, an unusually strong El Niño caused drought conditions in New York State (National Drought Mitigation Center 2016). This pattern was evident at the study sites as stream gauges reported lower flows than previous years (Figure 3). The altered flow and changing redox conditions through riparian and wetland areas may have allowed for increased methylation at the end of 2016. As these increases in MeHg production occurred at the end of the study I have no information on the effects of MeHg production in these systems as conditions improved through 2017 (NIDIS 2019).

4. Phase 2: Changes in Dissolved Organic Matter within a Honnedaga Lake Subwatershed

4.1 Methods

4.1.1 Study Site

For this phase, subsamples from CL1 and CR1 at Honnedaga Lake (phase 1) were collected for focused analysis of the characteristics of dissolved organic matter (Figure 2). A detailed description of these sites, sample collection and chemical analysis is provided in sections 4.1.1, 4.1.2 and 4.1.5.

4.1.2 Sample Selection

A subset of paired stream-water samples manually collected from CL1 and CR1 were selected to identify changes in DOM. These identified samples were two pre-treatment (September 2012), four short-term response immediately after the treatment (October and November 2013) and three longer -term response dates (July-September 2016). These samples cover the periods with the highest DOC, THg and MeHg concentrations (Table 8). The 2013 samples focus on the period just before and after treatment with $CaCO_3$ while samples collected during the 2016 field season saw the highest concentrations of MeHg at both reference and limed sites (Figure 16).

4.1.3 Sample Preparation

Samples were prepared for analysis on ESI-FTICR-MS following methods developed by Chen et al. (2017). DOM samples were concentrated by lyophilizing and redissolving material in 4 mL of Milli-Q water, resulting in a final estimated DOC concentration of 6 – 6.5 mmol C/L. These concentrated samples were then frozen in at -70 ^{o}C and shipped on ice to the Environmental Molecular Science Laboratory at the Pacific Northwest National Lab in Richland,

Washington. Once at the PNNL, a modified styrene divinyl benzene polymer (PPL) was used to perform a solid phase extraction and purify 1 mL of the concentrated DOM following a method similar to those previously described (Dittmar et al. 2008). In the modified method, milli-Q water was used to remove salts from the sample instead of concentrated HCl in order to preserve any Hg-DOM complexes. Assuming a 60% recovery from SPE, the extracted DOM was diluted with a 1:1 methanol:water solution to reach the target concentration of ~3.2 mmol/L as previously used for the identification of Hg:DOC complexes (Chen et al. 2017). An additional 2mL of each concentrated sample was lyophilized and dissolved in D_2O for ^{1}HNMR analysis.

4.1.4 Instrumental Analysis

My results were compared with previously developed MS libraries (Chen et al. 2017). ESI-FTICR-MS was applied to compare the number and identities of CHO, CHOS, CHON, CHOP, CHONS, CHONP, CHOSP, and CHONSP molecular formulae in limed and unlimed DOM extracts. The high-resolution mass spectra of DOM was measured on the in-house designed 21 Tesla FTICR mass spectrometer housed at the Environmental Molecular Sciences Laboratory (EMSL; Shaw et al. 2016). An Agilent heated electrospray ionization unit was used as the ion source with sample flow rate, spray voltage and heated capillary temperature set to 0.5uL/min, 3.5−3.6 kV and 275 °C, respectively. Spectra were acquired in both positive and in negative ionization mode (Tfaily et al. 2015, Chen et al. 2017) with resolving power set to $1*10^6$ at m/z 400 by automatic gain control (AGC). Peaks from 200 to 1000 *m/z* with a minimum signal-to-noise ratio of 3 were assigned molecular formulae. A molecular formula calculator Formularity software (Tolić et al. 2017) was used to generate formulas with mass measurement error < 1ppm, including only elements using C, H, O, N, P, and S and using formula building blocks of CH2, O and H2 homologous series to propagate formulas above m/z values of 500 Da.

In the case of multiple candidates, formulae with at least one oxygen atom, the lowest error, lowest number of non-oxygen heteroatoms, P <= 2, 3*P<=O and N <= 3 were selected. Peaks between spectra were aligned if the *m/z* values agreed within 0.5 ppm of each other. These assignments were also run individually to compare results for improved accuracy. Formulas were assigned based on previously described rules (Koch et al. 2007, Stubbins et al. 2010). Molecular ratios of O/C, H/C, and S/C for single formulas were used to construct *van Krevelen* diagrams ranking the relative abundance of the S-containing molecules (Kim et al. 2003, Wu et al. 2004, Mangal et al. 2016). Contour intensity plots of the C numbers, double bond equivalent (DBE) values, and relative intensities of S-containing molecules were constructed to evaluate the degree of unsaturation of S-containing molecules (Mangal et al. 2016).

4.1.5 Statistical Analysis

Statistical analyses of DOM data were performed with the software package Rv3.4.2 (R Core Team 2018). Van Krevelen diagrams were constructed for each selected water sample. These figures were used to identify changes in DOM quality following treatment. Areas of interest were identified and further broken down by the number of sulfur molecules contained within each formula assignment. The number of assigned formulas from these identified regions were regressed with concentration data from each sample to describe THg, MeHg, and MeHg/THg relationships with sulfur-containing hydrocarbons. In addition to the total number of S-containing functional groups in molecular formula assignments, regressions were also constructed separately for the 1-, 2- and 4-S formula assignments and the ratios between the number of assignments with each number of sulfur groups (1S:2S, 1S:4S, 2S:4S) were tested for significant relationships with THg, MeHg, and MeHg/THg (%MeHg). These analyses was also conducted using log transformed values.

4.2 Results

4.2.1 Dissolved Organic Matter Composition from Watershed Calcium Carbonate Treatment (CL1 and CR1)

The number of peaks detected in each sample was 5837.625±1185.52 (average ± standard deviation) with 5309.25±874.02 assigned peaks. Based on these assignments there is a notable shift in DOM composition following the $CaCO_3$ addition. The samples immediately following treatment had 14.29±3.28% of assigned peaks containing thiol functional groups. This value is twice the number from the September pre-treatment sample (8.6%) and the 2016 limed (9.66±0.81%) and reference samples (8.06±0.08%; summary Table 9).

Van Krevelen diagrams were constructed for each sample analyzed by FTICR-MS (Figure 17). These diagrams show that sulfur containing hydrocarbons with low oxygen and hydrogen to carbon ratios (O/C < 0.21; H/C < 1.6) changed dramatically between reference and treatment samples. Reference and pre-treatment diagrams had lower counts of single sulfur assignments in this region (Figure 18).

4.2.2 Linear Regression analysis

Analysis revealed that the number of unsaturated and condensed S-containing hydrocarbon assignments within the identified region (0.21 < O/C; 1.6 < H/C) was significantly related to THg ($m = 0.008$, $p = 0.048$, adj-$R^2 = 0.132$). This relationship became much stronger when excluding the first two treated samples ($m = 0.008$, $p < 0.001$, adj-$R^2 = 0.762$). Separating by site did not result in the same significant relationship for either treatment or reference. Further refining of the data revealed a significant positive relationship with 1-S assignments ($m = 0.008$, $p = 0.049$, adj-$R^2 = 0.244$) that improved when removing the first two treated samples ($m = 0.008$, $p < 0.001$, adj-$R^2 = 0.780$; Figure 20). There were also significant/marginally

significant negative relationship of THg with 2-S and 4-S assignments at the treatment site (m = -0.069, -0.239 p = 0.018, 0.063; respectively, Figure 20).

Natural log transformation of the ratio of sulfur assignments also resulted in significant/marginally significant negative relationships with the natural log of THg across all sites and periods (Figure 21; 2S:1S: m = -0.464, p < 0.001; 4S:1S: m = -0.253, p = 0.004; 4S:2S: m = -0.389, p = 0.061). This relationship was also significant for the log transformed 2S:1S ratio and THg at the treatment site (m = -0.42, p = 0.016). MeHg and %MeHg were not found to have significant linear or natural log transformed relationships with the unsaturated and condensed S-containing hydrocarbons.

4.3 Discussion

4.3.1 Dissolved organic matter response to watershed liming

In this section, I discuss findings related to the FTICR-MS analysis. A more in-depth discussion of the chemical response to watershed liming can be found in section 5.3.1 within the larger context for the Honnedaga Lake liming project. DOM and more specifically, thiol-containing organic matter has been previously identified as an important ligand in the transport and transformation of Hg (Haitzer et al. 2003, Skyllberg et al. 2003, Dittman and Driscoll 2009, Chen et al. 2017). My data suggests that 1-S (likely thiol)-containing hydrocarbons are an important part of this transport process as there was a significant relationship with the number of 1-S containing molecules found and THg concentrations. It is likely that these 1-S molecules are driving the overall trend. This 1-S group is also the heaviest, most aromatic (lowest H/C, O/C and S/C ratios Figure 22). While this class of molecule is important, it is not exclusively responsible for THg transport as evidenced by the two samples immediately following $CaCO_3$ treatment falling well outside of this pattern.

The significant negative relationship between the natural logs of 2:1 and 4:1 sulfur containing molecules and THg suggests that the 2- and 4-S molecules contribute to the production, aggregation and precipitation of HgS nanoparticles. These 2- and 4-S assignments also have lower molecular weight (Table 10) and lower aromaticity (higher H/C ratio) which has been shown to enhance methylation (Graham et al. 2012, 2013). These findings support the hypothesis from Millard et al. (2018) suggesting that watershed treatment with $CaCO_3$ mobilized larger, more aromatic DOM as the samples in the first month following treatment were enriched in the 1-S molecules.

5. Phase 3: New York State Survey

5.1 Methods

5.1.1 Fish sampling and processing

A subset of lakes across New York State previously sampled by Simonin et al. (2008) and lakes with multiple years of historical sampling were identified for study. Collections of fish samples began in 2014 and were concluded in 2016. Walleye (WE), Smallmouth Bass (SMB), Largemouth Bass (LMB) and Yellow Perch (YP) were targeted for collection because of their popularity among sport fishers and the relative abundance of previous Hg observations. Three alternative species, Brook Trout (ST), Chain Pickerel (CP) and Lake Trout (LT), were also collected from lakes where the target species were not available or abundant during the 2010s sampling. Working with partners at the NYS DEC, Cornell University and SUNY-Oneonta, fish samples were collected from 108 lakes from all regions of the state to evaluate spatial patterns and temporal trends (Figure 23).

Personnel from the NYS DEC, Cornell University, SUNY Oneonta and SU collected fish using standard sampling methods including gill netting, trap netting, minnow trapping, angling and electrofishing (when necessary), to obtain a minimum of 10 adult fish samples from targeted species. Collected fish were measured, weighed, individually placed in labeled, food grade plastic bags and kept on ice until analyzed at SU. Upon arrival, samples were prepared according to DEC methods (standard fillet; skin-on, bone-in, scales removed) and 524 subset fish plugs were also collected according to previously established procedures (Baker et al. 2004, Peterson et al. 2004). All samples were frozen at a temperature below -18^{o}C. Samples were freeze dried and milled before being analyzed on a Milestone Hg analyzer utilizing thermal decomposition, catalytic reduction, amalgamation, desorption, and atomic absorption

spectroscopy following U.S. EPA method 7473 (U.S. EPA 2007b). CCV and CCB samples were analyzed every ten samples, with duplicates and matrix spikes analyzed every 20 samples. Additional quality control samples (QCS) were analyzed at the beginning and end of each run (Appendix 3).

5.1.2 Water sampling and processing

In addition to fish, surface water samples were collected from 35 lakes from 2014 to 2016, using Teflon bottles and trace metal collection methods for analysis of total Hg (THg), MeHg, and plastic bottles and standard collection methods for SO_4^{2-}, NO_3^-, Cl^-, monomeric Al, ANC, pH, DOC, UV_{254} absorbance and chlorophyll *a*. These samples were placed on ice in the field and shipped to Syracuse University for analysis. Mercury samples were filtered using 0.45µm Millipore filters prior to analysis. Total Hg was analyzed by oxidation, reduction, purge and trap, desorption and cold-vapor atomic fluorescence spectrometry (CVAFS) according to U.S. EPA method 1631, revision E (U.S. EPA 2002). Continuing calibration verification (CCV) standards and blanks were analyzed every ten samples with additional duplicate and MS/MSD samples analyzed every twenty samples. An additional Quality Control Sample (QCS) was analyzed at the beginning and end of each analysis. The detection limit for this method is 0.2 ng/L. MeHg was analyzed by direct ethylation, purge and trap, desorption, and CVAFS following a modified U.S. EPA method 1630 (U.S. EPA 2007a).

Acid neutralizing capacity (ANC) and pH were analyzed in the SU laboratory using a Brinkmann Metrohm 716 DMS Titrino and 760 sample changer with a Ross General Purpose Sure-Flow pH electrode. Ion chromatography was used for analysis of SO_4^{2-}, NO_3^-, and Cl^-. Water samples were prepared for DOC analysis by filtering through a 1.5µm glass microfiber filter and analyzed using infrared detection following persulfate oxidation. UV_{254} was analyzed

by measuring absorbance at 254 nm in a standard spectrophotometer. Colorimetric methods were used to analyze for monomeric aluminum (Al_m) following chelation with pyrocatecholviolet. This same method was used to analyze for organic monomeric aluminum (Al_o) after the sample was passed through an ion exchange column. Inorganic monomeric aluminum (Al_i) was then calculated by subtracting Al_o from Al_m (McAvoy et al. 1992). Chlorophyll *a* samples were sent to the Upstate Freshwater Institute for analysis by concentrating chlorophyll containing particulate matter on 0.45 µm cellulose nitrate filters. The pigment was then extracted from the filter using acetone and measured using a standard spectrophotometer where a duplicate sample (±15 RPD) and standard reference material (±15% recovery) were analyzed every tenth sample for QA/QC.

5.1.3 Standardizing fish THg concentrations

In the earlier survey fish tissue Hg were characterized using a "DEC filet" (Department of Environmental Conservation). The DEC filet involves measuring mercury in fish tissue samples that represent a muscle filet, which includes fish skin and bones. In contrast, the recent survey involved collection of fish muscle tissue using a plug that does not include skin and bones. Because of these differences in analysis of fish muscle tissues, following laboratory analysis the percent difference of paired DEC fillet and plug samples were calculated. This comparison showed that DEC fillet results have an average low bias of 15% when compared to plugs. Note that there is no statistically significant difference in mercury concentrations between standard fillet and plugs (Knight et al. 2019). A conversion formula was developed to convert and compensate for the low bias of DEC fillet results (Montesdeoca et al., in progress). Four models were optimized to a Pearson correlation test and a Shapiro-Wilks residual normality test

using 90% of paired results for development and 10% for training. The following equation produced the best results for converting DEC fillet mercury results to plug results:

$$[Hg] = \frac{e^{\ln(length*weight)+0.14}}{length * weight}$$

where length and weight are for the whole fish.

The statistical software R (R Core Team 2018) was used to correct low bias of DEC fillet samples and standardize THg concentrations at lengths determined by the previous survey (YP - 229 mm, SMB - 356 mm, LMB - 356 mm, WE - 457 mm; Simonin et al. 2008a). Length standardization was achieved by fitting a linear model for each lake per year. Model residuals were then used to estimate the THg concentration of each sample at a standardized length. These length adjusted data were used to evaluate temporal trends, correlations with chemical and physical parameters, and conduct spatial analysis.

5.1.4 Statistical analysis on water chemistry and fish Hg concentrations in 2010s

Surface water chemistry data were evaluated for 18 lakes in the Northeast, 15 lakes in the Southeast and 2 lakes in the West region of New York State. A simple t-test was applied to compare water chemistry data among regions (i.e., Northeast vs. the Southeast) using lakes as replicates.

To study differences in Hg concentrations among the seven fish species in this study, a one-way ANOVA was applied to the standardized fish THg concentrations with lakes as replicates. For the four most commonly sampled species (YP, SMB, LMB and WE), Z-scores were calculated for the individual standard-length fish Hg concentrations. ArcMap (ESRI 2017)

was used to examine the spatial variation of the four most commonly sampled species across New York State.

A Pearson correlation test was used to examine the relationships between water chemical variables and fish Hg concentrations of the four common fish species. Linear regressions were used to investigate the effects of lake characteristics on water chemistry and fish Hg concentrations. Lake characteristics were acquired from Simonin et al. (2008a), including elevation (m), watershed area (ha), lake area (ha), shoreline (km), contiguous wetlands (ha), watershed wetland area (ha) and watershed wetland area (%). I also tested whether an existing outlet dam would impact water chemical variables and fish Hg of the four common species using one-way ANOVA with lakes as replicates.

5.1.5 Changes in water chemistry and fish Hg concentrations between 2000s and 2010s

The majority of the lakes in this study only have data from two sampling events. Paired t-tests were used to examine changes in water chemical variables and fish THg concentrations of each of the four common species state-wide and by region using lakes as replicates.

To examine if lake characteristics and changes in water chemistry (%) from 2000s to 2010s would explain the changes in fish THg concentrations in each of the four common species state-wide, linear regressions were used with lakes as replicates. Stepwise regression was also used to determine if any models would predict changes in fish THg concentrations for each species. ArcMap v10.6 was used to examine the percent change of THg concentrations in each species between the two collection periods. Kriging was again used for all species, and Inverse Distance Weighting (IDW) was used when kriging showed no variation across New York State (SMB, LMB).

To determine the number of lakes required to detect a change in fish THg concentrations after a decade, I calculated the sample size needed to detect a difference of 2-10% of the mean THg concentrations for each commonly species using paired t test:

$$N = 2 \times \frac{S^2}{d^2} \times (Z_{1-\alpha/2} + Z_{1-\beta})^2$$

where N is the sample size (number of lakes per sampling period), s^2 is the pooled variance, d is the detectable difference, Z is the critical value from the standardized normal distribution, $\alpha = 0.05$, and $\beta = 0.2$ (i.e., power is 0.8). Pooled variance can be calculated as:

$$s^2 = \frac{s_{\text{pre}}^2 + s_{\text{post}}^2}{n_{\text{pre}} + n_{\text{post}}}.$$

where s^2 is the variance for the treatment period and *n* is the sample size for the treatment period.

To investigate if a change in sampling intensity by region would impact the ability to detect trends, I calculated and compared the number of lakes required for each common species among the three regions to detect a 10% change in fish THg concentrations over a decade. Because the dataset only allowed detection of a temporal change between two points of time, it was not possible to examine the impact of sampling frequency. These statistical analyses were conducted with SAS 9.4 (SAS Institute Inc. 2013).

5.2 Results

5.2.1 General lake chemistry and standard-size fish Hg concentrations in 2010s

The 18 Northeast, 15 Southeast and 2 West lakes in NYS had water column THg (mean ± SD) of 0.86 ± 0.70 ng L^{-1}, MeHg of 0.06 ± 0.08 ng L^{-1} and %MeHg ((MeHg/THg)*100) of 7.4 ± 5.6. Additional statistics for individual lakes can be found in Table 11. The Northeast lakes had

a relatively lower ANC, pH and chlorophyll *a*, and a higher DOC, MeHg and THg than the Southeast lakes ($p \leq 0.05$). The two lakes in West region had similar water chemistry compared with Southeast lakes (Table 11).

The standard-size wet weight Hg concentration (mean ± SD) in 2010s was 0.43 ± 0.59 μg g^{-1} for YP (57 lakes), 0.22 ± 0.13 μg g^{-1} for ST (29 lakes), 0.58 ± 0.44 μg g^{-1} for LMB (22 lakes), 0.43 ± 0.21 μg g^{-1} for LMB (21 lakes), 0.67 ± 0.39 μg g^{-1} for WE (14 lakes), 0.81 ± 0.31 μg g^{-1} for CP (9 lakes) and 0.60 ± 0.61 μg g^{-1} for LT (8 lakes) in NYS (Figure 24). Chain Pickerel had higher Hg concentrations than WE, LT, LMB, LMB and YP; ST had the lowest Hg concentration among all species sampled ($p < 0.001$).

For the 43 resurveyed lakes, concentrations of Hg in YP muscle samples exceeded the U.S. EPA Fish Tissue Residue Criterion for MeHg of 0.30 μg g^{-1} in 71% of Northeast lakes, 8% of Southeast lakes and 0% of West lakes. Most lakes had Hg concentrations exceeding 0.30 μg g^{-1} for WE (10 out of 11 lakes), LMB (9 out of 12 lakes) and LMB (6 out of 10 lakes). Three Northeast lakes (North Lake for YP, Red Lake for WE and Hinckley Reservoir for LMB), one Southeast lake (Swinging Bridge Reservoir for WE) and one West lake (Rushford Lake for WE) had fish Hg concentrations that exceeded the US Food and Drug Administration action level of 1.0 μg g^{-1} (Figure 25).

5.2.2 Relationship between lake characteristics, lake chemistry and standard-size fish Hg concentrations in resurveyed lakes

Lake MeHg and THg concentrations were positively correlated with DOC, total monomeric Al and organic monomeric Al, and negatively correlated with pH and SO_4^{2-} (Table 12). Lakes with higher organic monomeric Al or higher MeHg had higher %MeHg ($p < 0.01$ for both).

Lake pH was negatively correlated with total monomeric Al (r^2 = -0.63, $p < 0.01$), DOC (r^2 = -0.35, p = 0.05), and positively correlated with ANC (r^2 = 0.39, p = 0.02) using a Pearson correlation test for all 35 lakes (Table 12).

Total Hg concentrations increased with increases in lake elevation (r^2 = 0.41, p = 0.03) (Figure 26). Two lakes (Rock Pond - 2.37 ng-Hg L^{-1}; Sunday Lake 3.64 ng-Hg L^{-1}) had relatively higher THg concentrations than the other lakes studied (0.22 – 1.55 ng L^{-1}). With data from those two lakes removed, the relationship of higher aqueous THg concentration at higher elevation remained significant (r^2 = 0.47, p = 0.01). %MeHg was positively correlated with shoreline length (r^2 = 0.44, p = 0.03) and % contiguous wetland relative to lake area (r^2 = 0.49, p = 0.04). Other lake characteristics such as watershed area, lake area and contiguous wetland area did not influence the lake chemistry ($p \geq 0.12$).

Mercury concentrations in standard-size YP (r^2 = 0.64, $p < 0.001$) increased with lake THg concentration, but this pattern was not evident for WE, LMB or SMB ($p \geq 0.29$). Lake MeHg concentration and % MeHg were not correlated with standard-size Hg concentrations for any of the four fish species ($p \geq 0.20$). Total monomeric Al, ANC, pH, and SO_4^{2-} were correlated with standard-size Hg concentrations for YP and LMB, but not for WE and LMB (Table 12).

As with water column THg, Hg concentrations in YP increased with lake elevation (r^2 = 0.60, $p < 0.001$), but not for WE, LMB and LMB ($p \geq 0.59$). Lakes with a larger percentage of contiguous wetland relative to lake area had higher standard-size Hg concentrations for WE (r^2 = 0.88, p = 0.02), but not for YP, LMB and LMB ($p \geq 0.27$). Other lake characteristics such as watershed area, lake area, shoreline length and the presence of an outlet dam did not influence fish Hg concentrations ($p \geq 0.14$) in the studied lakes.

5.2.3 Changes in resurveyed lake chemistry between 2000s and 2010s

For all resurvey lakes, concentrations of THg decreased 42% ($p < 0.01$), MeHg decreased 74% ($p < 0.01$), SO_4^{2-} decreased 77% ($p < 0.01$), NO_3^- decreased 39% ($p = 0.02$), and total monomeric Al decreased 25% ($p = 0.01$) from 2000s to 2010s. Lake ANC increased 6% ($p = 0.02$), DOC increased 22% ($p = 0.04$). The pH ($p = 0.13$), chlorophyll a ($p = 0.9$) and Cl ($p = 0.18$) were similar between the two sampling periods. There was no statistically significant change in the %MeHg values between the two sampling periods.

In both Northeast and South regions, lake THg ($p < 0.001$ for both), MeHg ($p = 0.004$ and 0.01) and SO_4^{2-} ($p < 0.001$ for both) decreased from 2000s to 2010s. While, lake pH ($p = 0.02$) and NO_3^- ($p = 0.05$) decreased. DOC ($p = 0.002$) increased only in Northeast lakes. Total monomeric Al ($p = 0.003$) decreased in only Southeast lakes.

5.2.4 Changes in standard-size fish Hg concentrations between 2000s and 2010s

Concentrations of Hg in LMB decreased 21% from 2000s to 2010s ($p = 0.07$). Standard-size Hg concentrations were similar between 2000s and 2010s for YP, WE and LMB ($p \geq 0.49$) using all paired lakes. Temporal changes in fish Hg concentrations varied by lake and species (Table 13). In lakes sampled for YP (n=36) and WE (n=12), fish Hg concentrations increased in about as many study lakes (53% and 58%, respectively) as lakes in which values decreased (47% and 42 %, respectively) from 2000s to 2010s. In contrast, lakes sampled for SMB (n=13) and LMB (n=10) decreased in a much larger fraction of the lakes surveyed (77% and 70%, respectively).

For lakes with ≥ 2 fish species sampled, seven out of 18 lakes showed the same temporal patterns of change in Hg concentrations in collected fish species. Two lakes (Hinckley Reservoir in the Northeast and Goodyear Lake in Southeast) had increases in fish Hg concentrations and

five (Red Lake in the Northeast; East Sidney Reservoir, Fort Pond, Onteora Lake and Rio Reservoir in the Southeast) showed decreases in fish Hg concentrations.

Dividing the lakes into the three regions (Northeast, Southeast, West), Hg concentrations in YP from lakes in the West increased 29% from 2000s to 2010s (average of five lakes, p = 0.04). Concentrations of Hg in LMB from Southeast lakes decreased 41% from 2000s to 2010s (average of six lakes, p = 0.02). Concentrations of Hg in other species from other regions did not change significantly ($p \geq 0.15$).

5.2.5 Changes in standard-size fish Hg concentrations between 2000s and 2010s explained by lake characteristics and water chemistry

Changes in Hg concentration in WE over time were positively correlated with lake elevation (r^2 = 0.67, p = 0.04). Changes in Hg concentration in LMB were also positively correlated with contiguous wetland area (r^2 = 0.86, p = 0.01). No empirical models could predict the changes in fish Hg concentrations from the 2000s to the 2010s using a stepwise regression of lake characteristics and changes in water chemistry.

The changes in fish Hg concentrations from 2000s to 2010s might be explained by the changes in lake chemistry over the same sampling period. In a Spearman rank test between percent changes in fish Hg concentrations for each species and the differences in chemical variables (ratio strength for difference in pH and percent difference for other variables), I found that changes in lake ANC (r^2 = 0.87, p < 0.001), and pH (r^2 = 0.74, p = 0.02) were positively correlated with changes in Hg concentrations in WE. Lake chlorophyll *a* was negatively correlated with changes in Hg concentrations in LMB (r^2 = -0.58, p = 0.04). Changes in water THg concentration (r^2 = -0.93, p < 0.001) was negatively correlated with changes in Hg concentrations in LMB.

5.3 Discussion

5.3.1 Variation in Hg in NYS lakes

In 2011, the Mercury and Air Toxics Standard (MATS) addressed the need for further emissions reductions by requiring a 90% reduction in Hg emissions from energy generation facilities (U.S. EPA 2017). Coal-fired power plants are the largest source of U.S. Hg emissions accounting for approximately 48% of Hg emissions in 2015 (Streets et al. 2018). MATS and earlier air quality management regulations have resulted in decreases in Hg emissions (Zhang et al. 2016, Streets et al. 2018) and decreases in atmospheric Hg concentrations (Zhou et al. 2017) and atmospheric Hg deposition (Gerson et al. 2016, Ye et al. 2019) in the northeastern U.S.

Freshwater resources in NYS can be sensitive to atmospheric Hg deposition (Driscoll et al. 2007b). All regions of the state show lakes with elevated concentrations of Hg in fish, with the highest concentrations generally found in samples collected from the Adirondack or Catskill park regions (Simonin et al. 2008a). Observations of YP, the most widely sampled fish species, indicate that the lakes within the Adirondack Park contain fish with the highest concentrations of Hg (Figure 25). Indeed, of the 106 lakes with specific fish consumption advisories for Hg in New York State, 62 occur in the Adirondack region. Moreover, there are regional advisories on fish consumption for the Adirondacks and Catskills (NYS DOH 2018). Even in regions of the state where fish Hg concentrations are generally lower, many of the popular gamefish remain above the 0.3 μg g^{-1} guideline (Figure 23).

With the majority of standard-length fish Hg above the 0.3 μg g^{-1} criterion, additional efforts are needed to reduce fish Hg concentrations. My analysis shows that despite decreases in emissions and atmospheric Hg deposition, there has not been a systematic decrease in fish Hg in NYS over the past decade. Rather, fish Hg in many lakes has increased relative to measurements

from the survey of Simonin et al. (2008). This pattern is consistent with findings from the Laurentian Great Lakes where decreases in atmospheric Hg were significantly correlated with decreases in fish Hg for all the lakes until 2010, when the patterns for Michigan, Erie and Ontario changed from decreasing to no change or increasing concentrations of fish Hg (Zhou et al. 2017). Like Lakes Michigan, Erie and Ontario, the major driver of fish Hg trends in NYS inland waters may be shifting away from regional Hg emissions towards the effects of legacy Hg inputs or increasing global Hg emissions, changes in nutrient status, invasive species, climate change and/or global Hg emissions.

I observed marked decreases in water column THg and MeHg concentrations between the two surveys. Decreases in atmospheric Hg deposition could explain this pattern of decreases in water column concentrations. Concentrations of THg and SO_4^{2-} alone were unable to explain the spatial variation in MeHg and %MeHg. While THg and SO_4^{2-} are likely important drivers of MeHg, I found that only length of shoreline and wetland area were significantly correlated with MeHg concentrations in study lakes. This pattern suggests that landscape features where reducing conditions occur control the extent and rate of methylation and MeHg concentrations, similar to findings from other studies (Mitchell and Gilmour 2008, Skyllberg 2008, Burns et al. 2012).

Many of the spatial correlations of biophysical and chemical factors with fish Hg found by Simonin et al. (2008) were not evident in this resurvey. In my resurvey, only concentrations of THg in lake water were significantly correlated with Hg in YP, but not other fish species. The response of fish to decreases in atmospheric Hg deposition may be influenced by recovery from the impacts of acid deposition, particularly in the Adirondacks, (Driscoll et al. 2001, Jeffries et al. 2003). Increases in fish Hg and aquatic concentrations of DOC have been associated with

recovery from acidification in northeastern North America and northern Europe (Monteith et al. 2007, Hongve et al. 2012, Åkerblom et al. 2012, Driscoll et al. 2013, Millard et al. 2018). This pattern is consistent with my findings where regions more highly impacted by acid deposition (Northeast region) have exhibited an increase in fish Hg which is coincident with increases in DOC.

Not only could chemical recovery from acid deposition alter fish Hg, but food webs could also be changing as acid-sensitive species recolonize impacted aquatic ecosystems. A longer food chain could result in a shift to higher trophic positions for sportfish and a subsequent increase in bioaccumulation (Ward et al. 2010). Conversely, increased nutrient inputs could result in biodilution or growth dilution of fish Hg through increases in aquatic productivity or accelerated individual growth (Riva-Murray et al. 2011, Kolka et al. 2019). Additional alterations to aquatic food webs such as stocking, or the introduction and/or management of invasive species can also have large impacts on food web dynamics and the biological cycling of Hg and may help explain why disparate trends in Hg concentrations were observed for different fish species within the same lake (Taylor et al. 2019). Community composition and structure play an important role for individual lake responses (Todorova et al. 2015) and further complicate statewide and regional trends.

The impacts of climate change contribute an additional level of uncertainty. Past and projected future climate for the Northeast have and will result in increases in air temperature and precipitation quantity (Climate Science Special Report 2017). A warmer and wetter climate in NYS could enhance primary production, lengthen the growing season, increase drought frequency, intensify precipitation and alter foodwebs (Hayhoe et al. 2007a), which could impact deposition, transport, methylation and bioaccumulation of Hg. Within this survey YP in the

West region showed almost a 30% increase from previous observations, while no change was detected for WE and LMB (potentially due to lower power to detect changes). These changes in the West region may be a result of climate driven impacts evidenced by an increased incidence of algal blooms in this region (Halfman 2017). This could result in a shift in autochthonous vs. allochthonous organic matter input which recent studies in boreal (ca. 59-60^{o}N; Bravo et al. 2017) as well as moist subtropical (ca. 26-28^{o}N; Jiang et al. 2018) lakes have shown a positive correlation between autochtonous organic matter and Hg methylation in sediment. Conversely, this could also result in lower fish Hg as a result of growth dilution (Essington and Houser 2003, Kolka et al. 2019) or biodilution, as has been reported in phytoplankton and zooplankton (Pickhardt et al. 2002, Chen and Folt 2005b).

Beyond these ecosystem complexities, an important consideration in the lack of fish Hg response to decreases in domestic Hg emission and local deposition may simply be time. A decade may be too short a period for adult fish to respond to additional decreases in atmospheric Hg deposition.

5.3.2 Number of lakes required to detect changes in standard-size fish Hg concentrations after a decade

The number of lakes required to detect a change between two sampling points would decrease if the rate of change in fish Hg concentration per decade was greater. Comparison among species of the number of lakes required to detect a given change in fish Hg showed that LMB and WE would require the greatest number of monitored lakes followed by LMB and YP (Figure 27). For example, it would require 18 lakes for LMB, 12 lakes for WE, 10 lakes for LMB and only 5 lakes for YP to detect a 4% change after a decade.

In order to detect changes in fish Hg, lakes in the West will require a greater number of sampled lakes in order to detect a change of 10% in fish Hg per decade than in the Northeast and Southeast (Figure 28). Southeast lakes required a continuation of current sampling efforts to detect a 10% change in fish Hg per decade for each of the four most commonly sampled fish species. In contrast the number of lakes required to detect a 10% change in fish Hg varied greatly for different fish species in the Northeast and West regions, with YP needing little additional sampling effort while targeting WE would require significant increases in sampling effort in both regions. Largemouth and SMB would also need significantly increased sampling efforts in the Northeast and West respectively.

5.3.3 Implications for continued monitoring of Fish Hg in NYS

Given the major pathway of human exposure to MeHg is by consumption of contaminated fish (Driscoll et al. 2013) and the elevated and increasing concentrations of Hg in NYS freshwater fish, monitoring of fish Hg in NYS should continue. With over 4000 lakes and many kilometers of rivers in the state (Simonin et al. 2008a) it is not possible to monitor all surface waters. However, a well-designed program could be used to monitor the regions defined by Simonin et al. (2008) or the ecological zones (ecozones) of New York State. For the current three regions, our power analysis shows that continuing previous sampling efforts of YP could detect a 10% change over a decade without increasing the monitoring effort.

Our analysis indicates that under the current three-region design, a lake monitoring program targeting 30 lakes (15 in the Adirondack State Park and 15 in the remainder of the state) would have a reasonable power to detect change in fish Hg concentrations (Figure 27). Sites outside the Adirondacks should likely include 1) lakes in the Catskills; 2) lakes of high economic, recreational and/or cultural importance such as the Finger Lakes, Oneida Lake, and Lake

Chautauqua; and 3) reservoirs (Rushford). Lakes should be selected based on the length of existing data records and those with recent fish Hg concentrations that exceed consumption guidelines.

In order to maintain a high statistical power, the majority of these lakes should be sampled at a five-year time interval, with a few lakes sampled more intensely in order to assess interannual variability. These more frequently sampled lakes may also be good candidates for more intensive analysis of lower trophic position species such as invertivorous fish species (e.g., dace species) or young of the year YP. Collecting smaller invertivorous species and/or young of the year would provide an opportunity to evaluate interannual variability and should exhibit a more rapid response to future controls on anthropogenic Hg emissions.

An alternative to this three-region model, would be sampling within the 12 ecozones within NYS (Figure 29; Will et al. 1982, Dickinson 1983). While this approach would likely require a significant increase in sampling effort, it would provide a much better understanding of fish Hg response to policy decisions. If a similar program for stream and river species were to be adopted, it might be possible to cover the majority of ecozones within NYS without too much additional effort.

6. Conclusions

6.1 Phase 1

My observations indicate a clear difference in impacts to the mobilization and transport of Hg among the two strategies used to amend streams with $CaCO_3$. Watershed scale applications have been shown to be effective for long-term improvements in soil and water quality, but my work demonstrates this approach can result in elevated concentrations and loading of DOC and THg. My data present no evidence for an increase in surface water MeHg or macroinvertebrate MeHg bioaccumulation within three years after liming; however, this condition could change over a longer period, and it could differ in other landscapes with greater potential for methylation, and uptake and bioconcentration of MeHg at the base of the food web.

Direct stream applications of $CaCO_3$ did not cause the same sustained mobilization of THg and other solutes as shown by watershed application. Responses to this treatment were rapid (<96 hrs) with reduced mobilization following repeated applications. Proximity and size of upstream wetlands appear to play a role in interannual variability, providing relatively greater background loads of THg and MeHg to downstream areas. The presence of wetlands did not explain any observed differences in treatment effect, however only one study site had a strong wetland influence. My results indicate that the timing of addition may have an impact on aquatic concentrations and bioaccumulation; winter addition could be less likely to cause biotic MeHg increases than applications occurring during the growing season.

These results also suggest a complimentary impact of recovery from acid deposition and climate change. In the final year of this study, concentrations of MeHg and the proportion of MeHg (%MeHg) increased across the majority of our study sites. This was likely caused by a strong regional drought and indicates that an intensified hydrological cycle could cause increases

in methylation. This along with other potential climate change impacts and the ongoing recovery from acid deposition could cause increased aquatic and biotic concentrations of mercury.

6.2 Phase 2

Sulfur containing unsaturated and condensed hydrocarbons are a major contributor to the transport of Hg from terrestrial to aquatic ecosystems. The number of these molecules present in samples had a significant positive relationship with total mercury concentrations across all sites and treatment periods. Following treatment with $CaCO_3$, elevated THg concentrations appear to be largely driven by larger, more aromatic organic molecules with only one sulfur containing (likely thiol) functional group. These molecules have been shown to inhibit the formation of HgS nanoparticles, potentially enhancing downstream methylation of Hg.

These large hydrocarbons may also be used preferentially by sulfate-reducing bacteria. In addition to increasing the number of monosulfuric hydrocarbons, treatment with $CaCO_3$ appears to reduce the number of polysulfuric hydrocarbons, which may result in a reduction in microbial processing and Hg uptake in terrestrial or riparian ecosystems. In areas recovering from acidic deposition, treatment with $CaCO_3$ may result in exchanging a long-term, gradual release of soil Hg from a natural recovery of pH, for a short-term pulse.

6.3 Phase 3

Across NYS, concentrations of Hg and MeHg in water largely decreased, however there is no evidence to suggest these improvements have also occurred in fish. Species- and region-specific trends varied widely, despite decreases in emissions and atmospheric Hg deposition, suggesting a diversification of important drivers. Fish Hg in NYS remains an ongoing problem as many popular gamefish remain above guideline consumption values, particularly in the Adirondack and Catskill regions.

This pattern suggests the major driver of fish Hg trends may be shifting from reductions in regional emissions to a variety of other factors. This shift has altered the relationships identified in previous fish Hg surveys making it more difficult to assess the effectiveness of U.S. policy and indicating the need of a fish Hg monitoring program. A well designed program could examine trends in the three regions identified by Simonin et al. (2008a), or the 12 ecoregions of NYS. The ability of this program to identify differences between regions could be essential for determining the different drivers of future mercury dynamics. For example, elevated temperatures and an intensified hydrological cycle suggests a potential shift in Hg drivers that contrasts with the Adirondack and Catskill state parks where legacy impacts of acidic deposition remain important to fish Hg. A better understanding of mercury drivers across NYS will inform future policy decisions and protect ecosystem and human health.

7. Synthesis and Integration

The fate of Hg in aquatic ecosystems depends on a complex set of chemical and ecological characteristics. Under changing climate regimes and legacy acidic deposition impacts the drivers of mercury fate appear are likely to become more varied in the future. While there is no consensus on the relative importance of autochthonous and allochthonous DOM to Hg methylation, my work suggests that larger, more aromatic sulfur-containing DOC play an important role in the transport of Hg from terrestrial to aquatic. My data suggests that watershed $CaCO_3$ treatment at Honnedaga Lake caused a release of this type of DOM from soil pools across the watershed, while channel applications mobilized DOM from a significantly reduced area.

Despite these increases in Hg and DOM, there were very limited impacts to biological accumulation. Only the winter application resulted in temporary elevated macroinvertebrate Hg concentrations which suggests the timing of DOM and Hg release played an important role in bioaccumulation. The lack of a biological response to other treatments could be due to Hg being bound to less bioavailable DOM when alternative carbon sources are available or being transported quickly through these oxic headwater streams. The results from Honnedaga Lake demonstrate potential for downstream impacts of watershed $CaCO_3$ additions on fish mercury. This is of particular concern in the Adirondack and Catskill regions where fish Hg remains elevated. By targeting streams and using channel application where appropriate it is possible to improve water quality while mitigating downstream concerns.

Addressing these downstream concerns would be important for making appropriate management decisions. Biological responses tend to lag behind chemical changes, especially in longer lived, economically important sport fish. Continued monitoring of mercury

concentrations in the lake resident Brook Trout at Honnedaga Lake could provide important information about the downstream biological response to these watershed management strategies.

Additionally, understanding the mechanism by which mercury is removed from soils could be critically important for predicting the impact of climate change and recovery from acidic deposition. In all three of my phases, only shoreline length and wetland area showed a significant relationship with %MeHg. Examination of DOM from soils and soil extracts could further characterize DOM important to the Hg transport and methylation process. This work would be important to expand beyond the Adirondack and Catskill regions of NYS, as an intensified hydrological cycle due to climate change could result in increased methylation periods across the region.

8. Figures

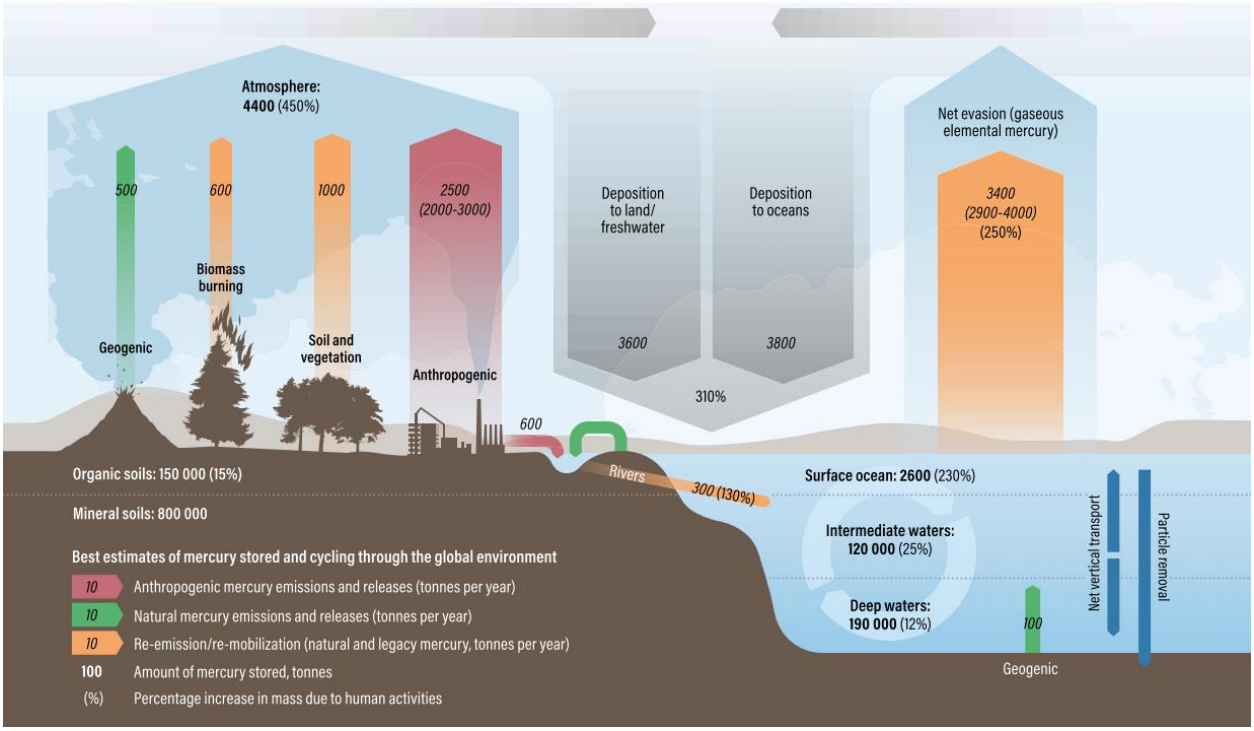

Figure 1: Estimates of global mercury cycling from the United Nations Global Mercury Assessment 2018 (UN Environment Programme Chemicals and Health Branch 2019).

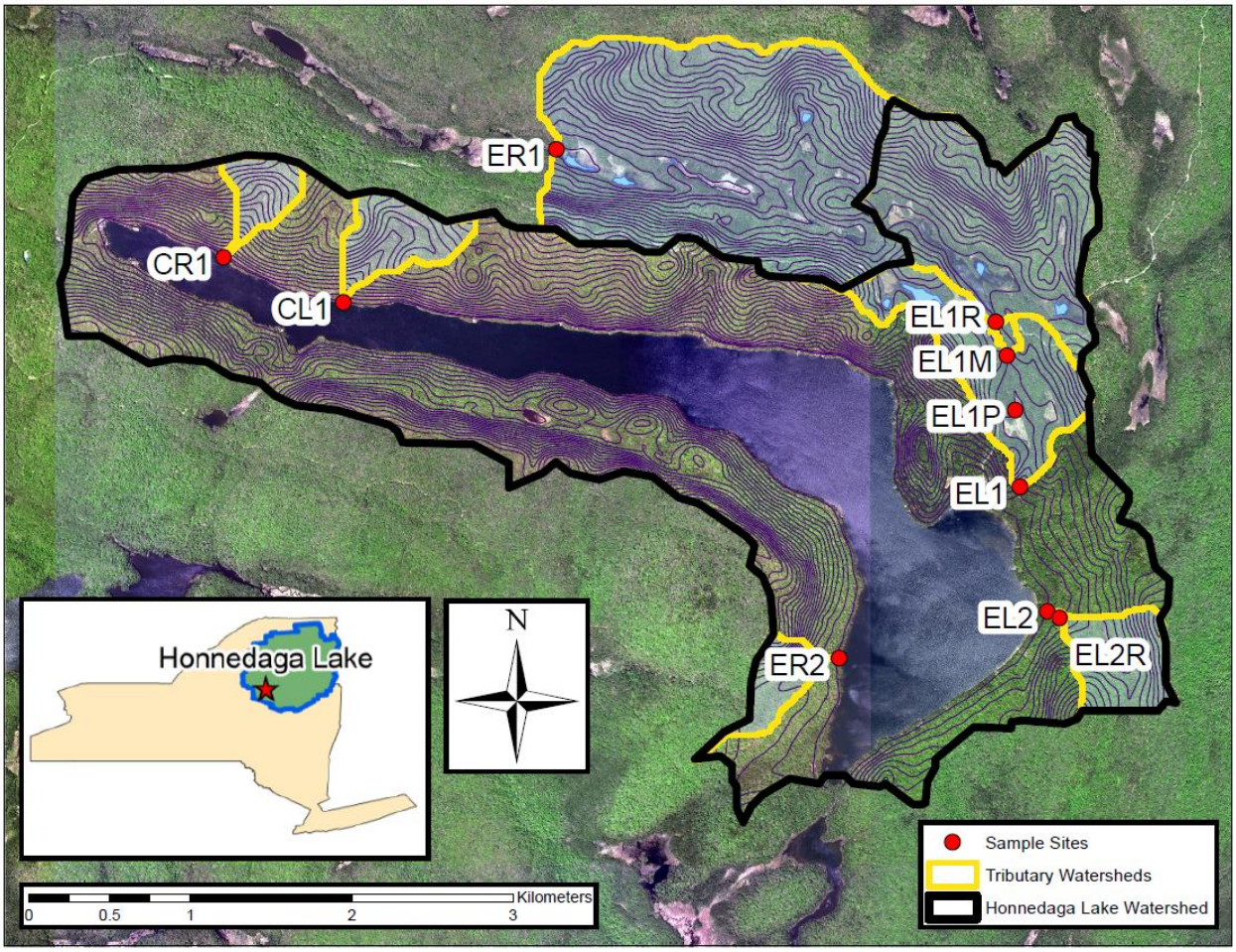

Figure 2: Map of Honnedaga Lake and the limed and reference watersheds. Inset map of New York State shows Adirondack Park in green, and the location Honnedaga Lake within the Park. This area of New York State is at a high elevation relative to the surrounding region. Each of the EL sites have a reference site above stream of the lime application (EL#R) and EL1 has two additional treatment site directly below the point of lime addition (EL1M) and directly above a wetland area (EL1P).

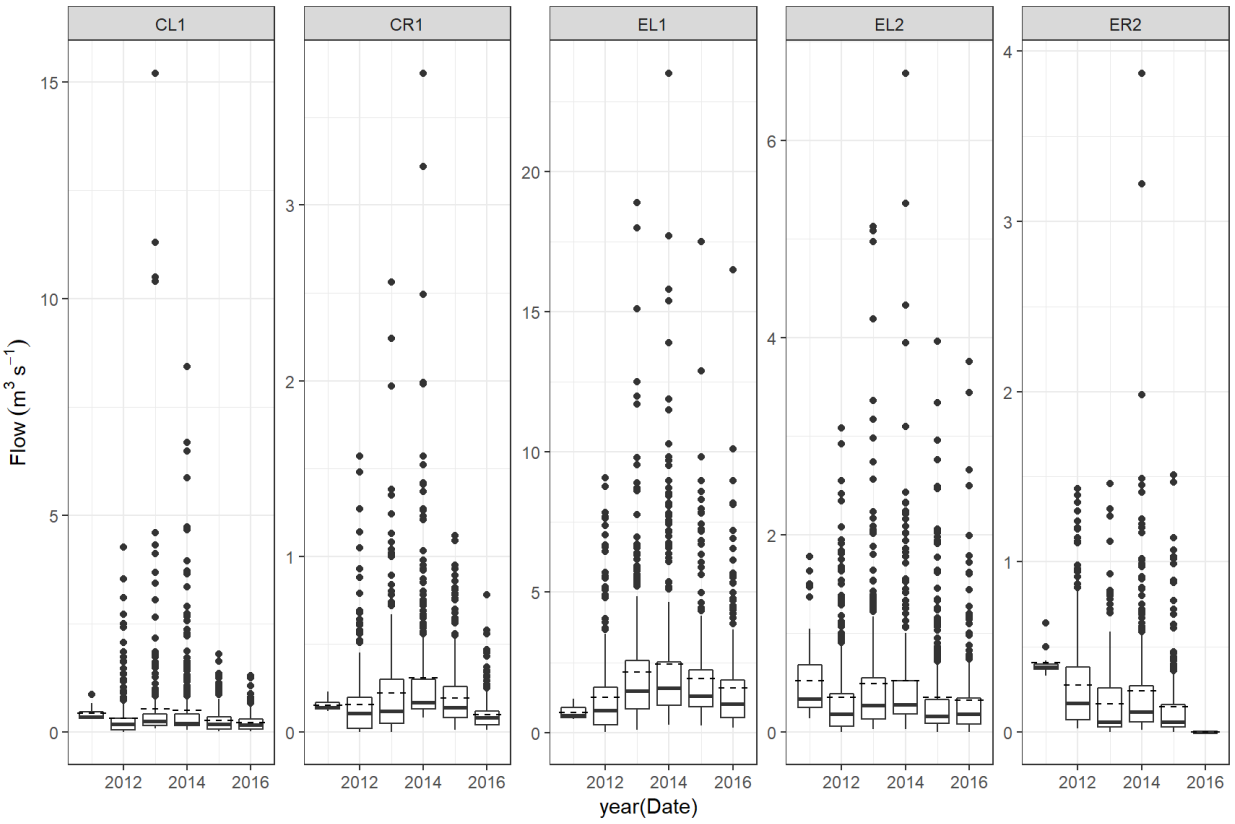

Figure 3: Annual daily flows measured at the outlet of each tributary to Honnedaga Lake with a USGS gauging station. No gauging station was present on ER1. Boxplots show the median as a solid line, the mean as a hashed line, the first and third quantile within the box and the whiskers extend to 1.5* the interquartile range. Individual points are outliers. CL1 = Chronically acidic, Limed site 1; CR1 = Chronically acidic, Reference site 1; EL1 = Episodically acidic, Limed site 1; EL2=Episodically acidic, Limed site 2; ER2 = Episodically acidic, Reference site 2.

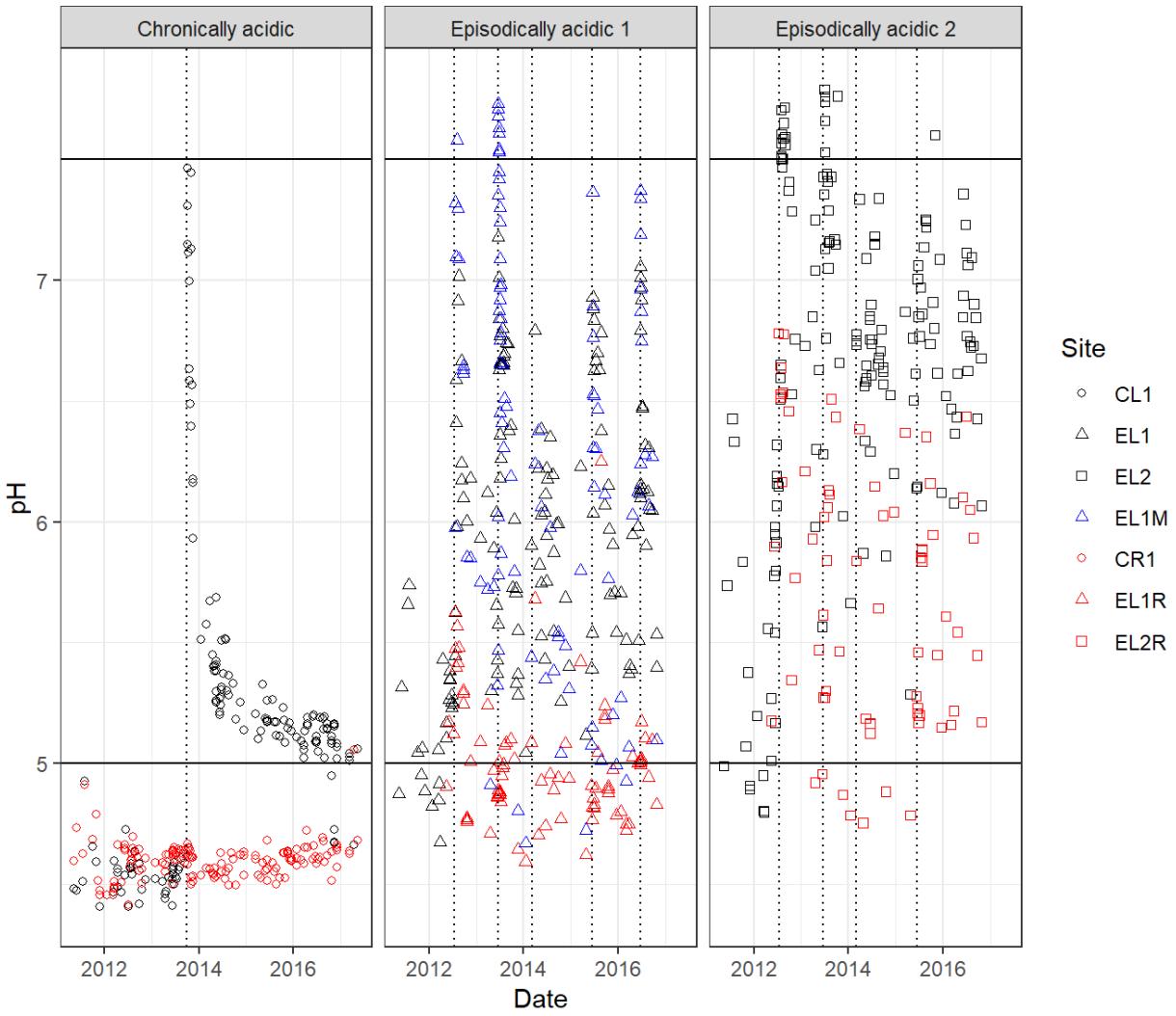

Figure 4: Time series of pH at each tributary except ER1 and ER2 to Honnedaga Lake. Watershed calcium carbonate amendment was effective at maintaining pH levels above 5 for several years, but became episodically acidic during the drought at the end of the study. Direct stream additions were effective for maintaining a pH above 5 at sites sufficiently downstream of addition points. Black points are treated sites, red points are reference sites and blue is physical located in between treatment and reference locations.

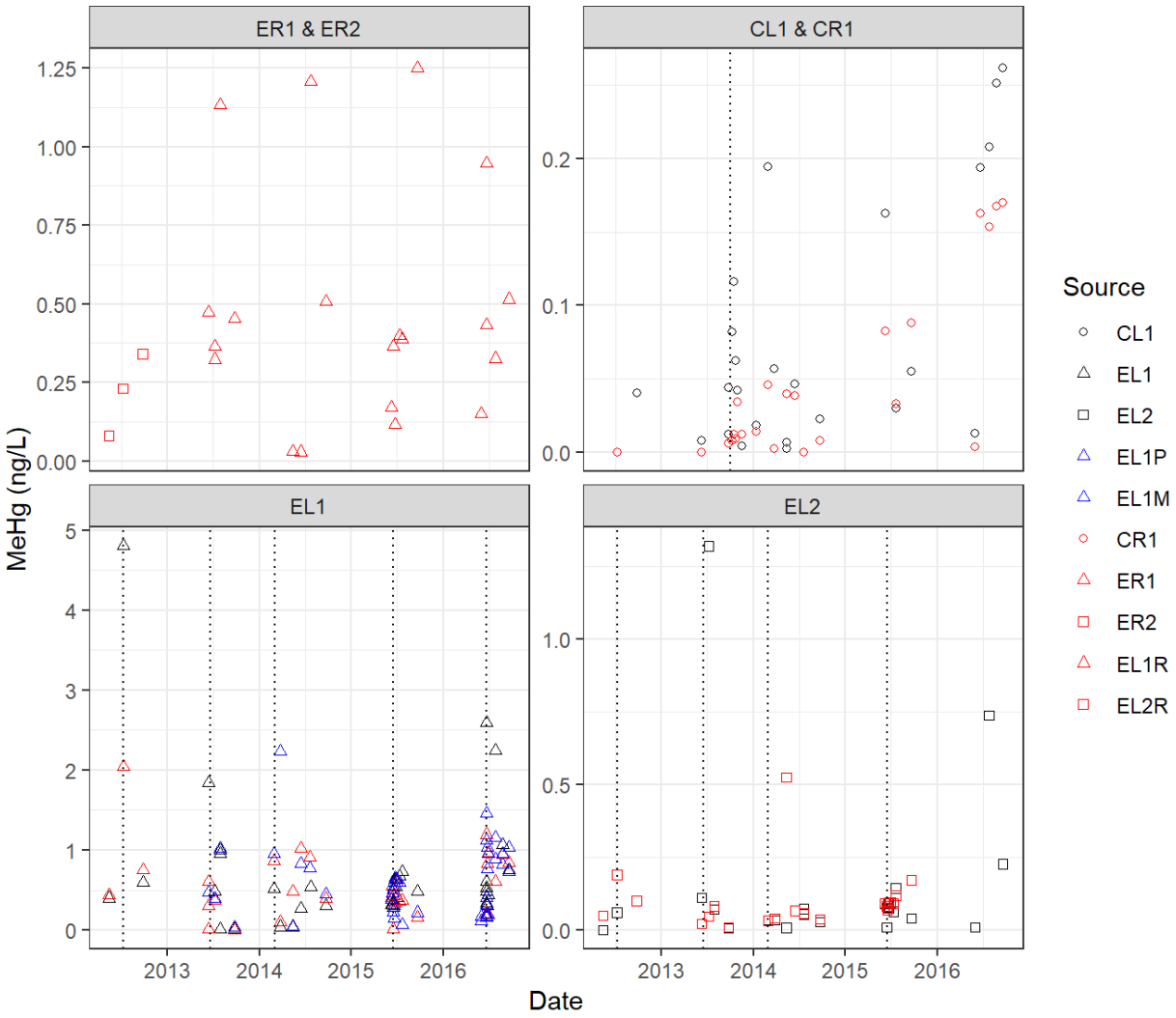

Figure 5: Concentrations of MeHg over time in Honnedaga Lake tributaries. Concentrations at CL1 were not found to be significantly different from those at CR1 after the first 6 months following treatment. Black points are treated sites, red points are reference sites and blue is physical located in between treatment and reference locations.

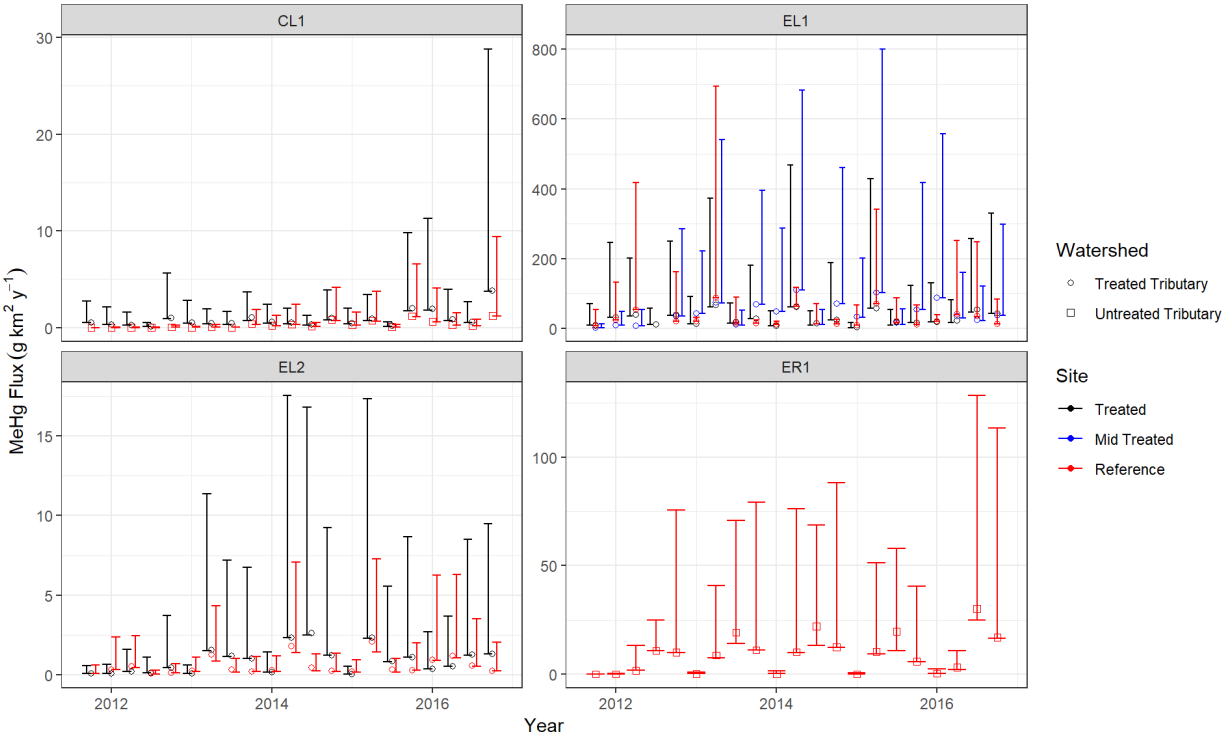

Figure 6: Seasonal fluxes calculated for all tributaries to Honnedaga Lake. CL1 and CR1 had large increases in flux in 2016, and EL2 experienced a significant increase relative to EL2R in the summer of 2014.

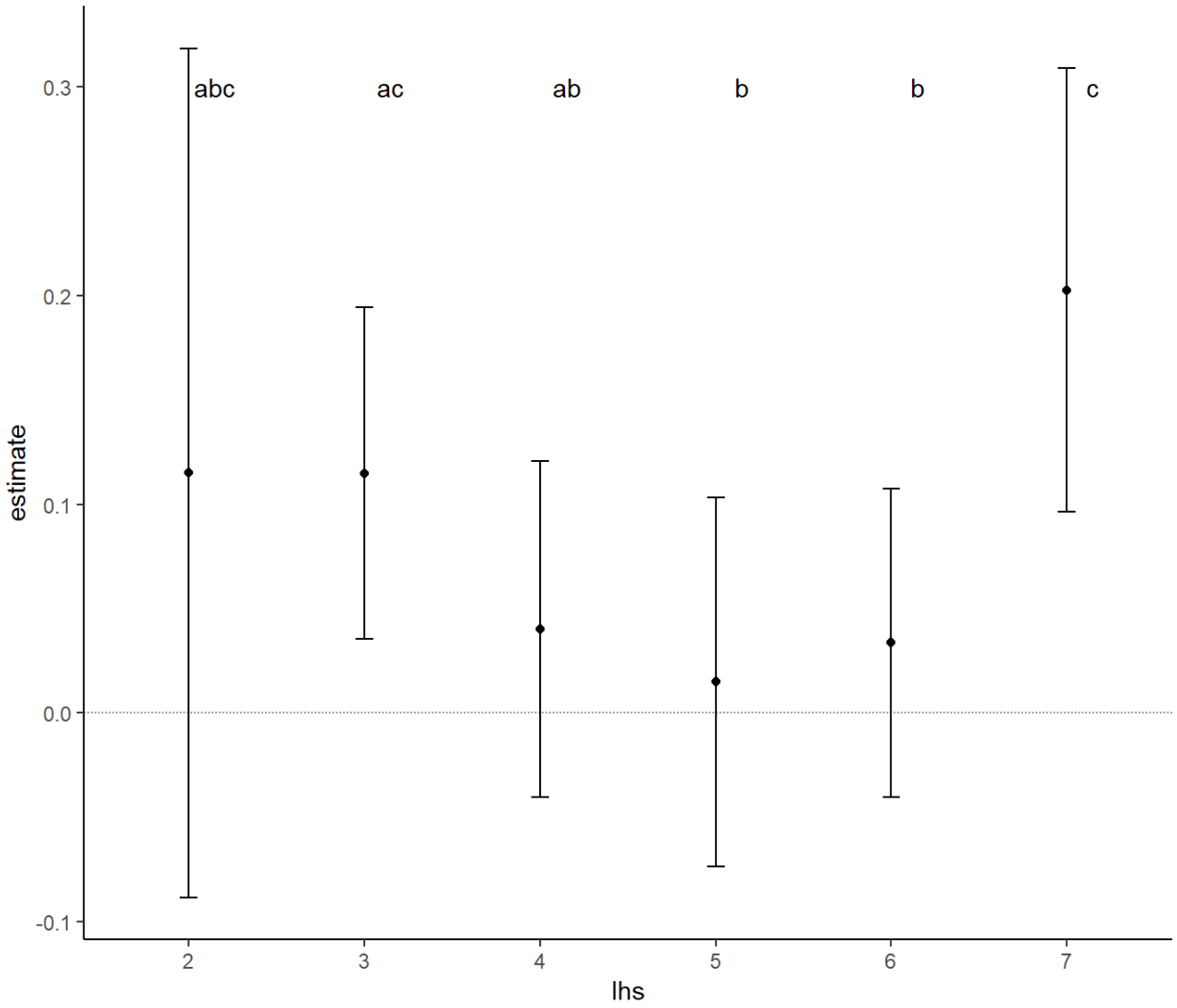

Figure 7: Statistically significant differences between UV_{254} at EL1 and EL1R for different sampling periods.

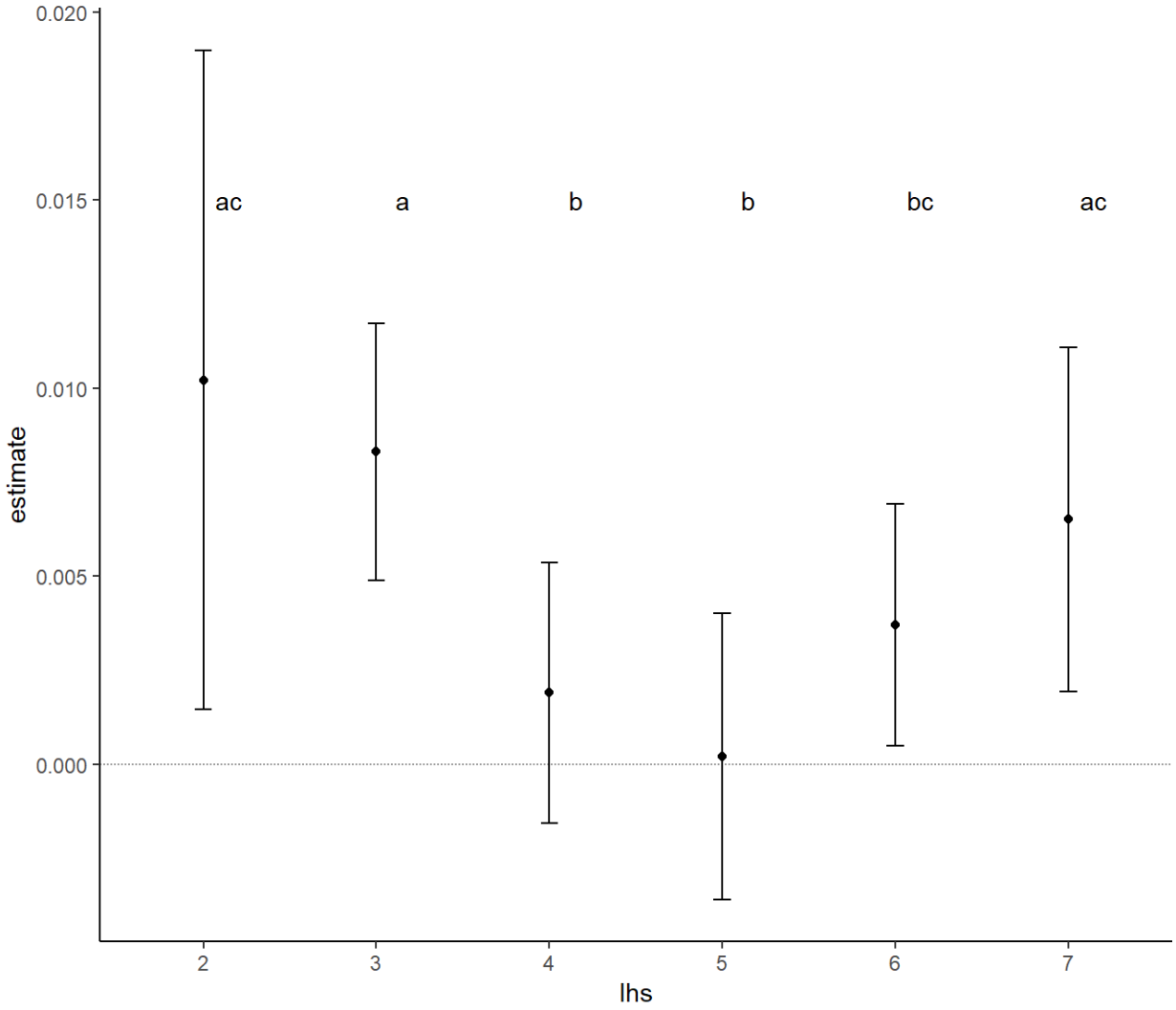

Figure 8: Statistically significant differences in SUVA between EL1 and EL1R at Honnedaga Lake watershed for different sampling periods.

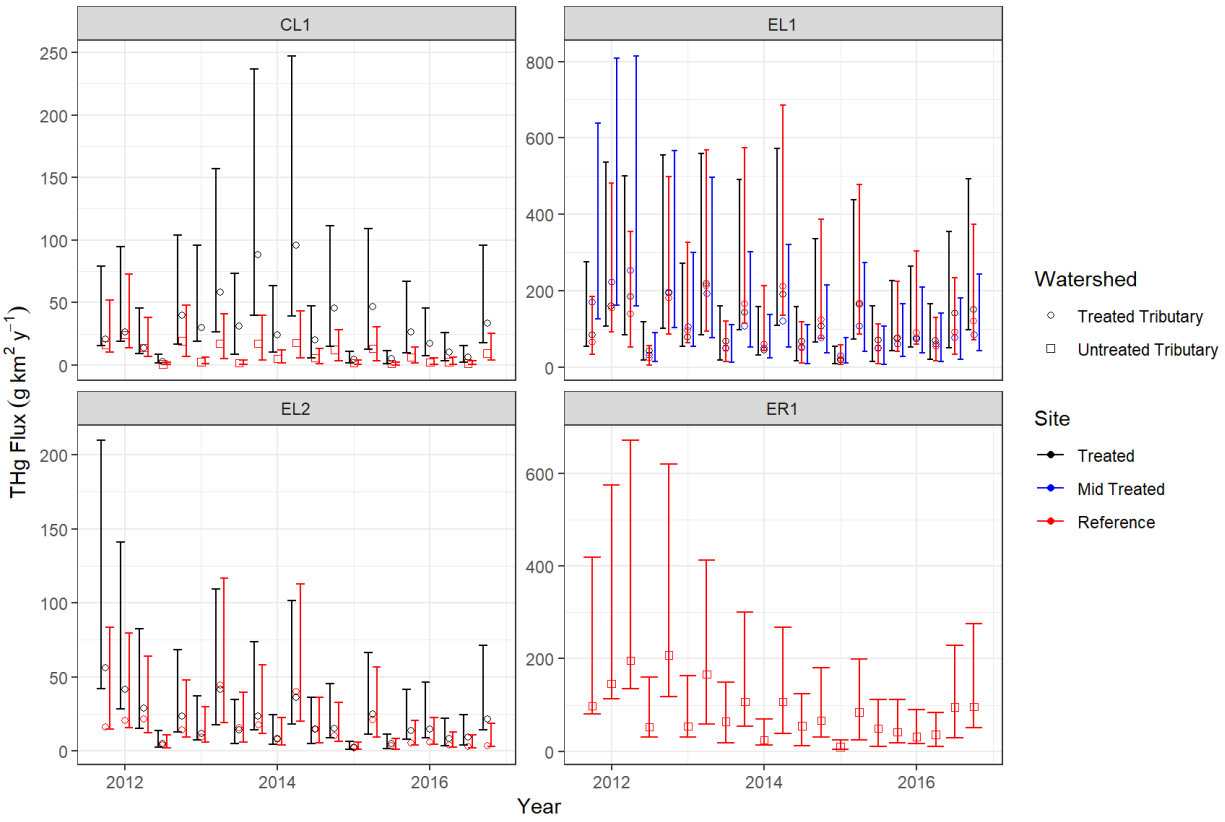

Figure 9: Seasonal loads of THg calculated for tributaries to Honnedaga Lake with sufficient Hg data. Only CL1 and CR1 have significant differences following calcium amendment. EL1 and ER1 have much higher loads than the other study sites due the larger wetland influence in these tributaries.

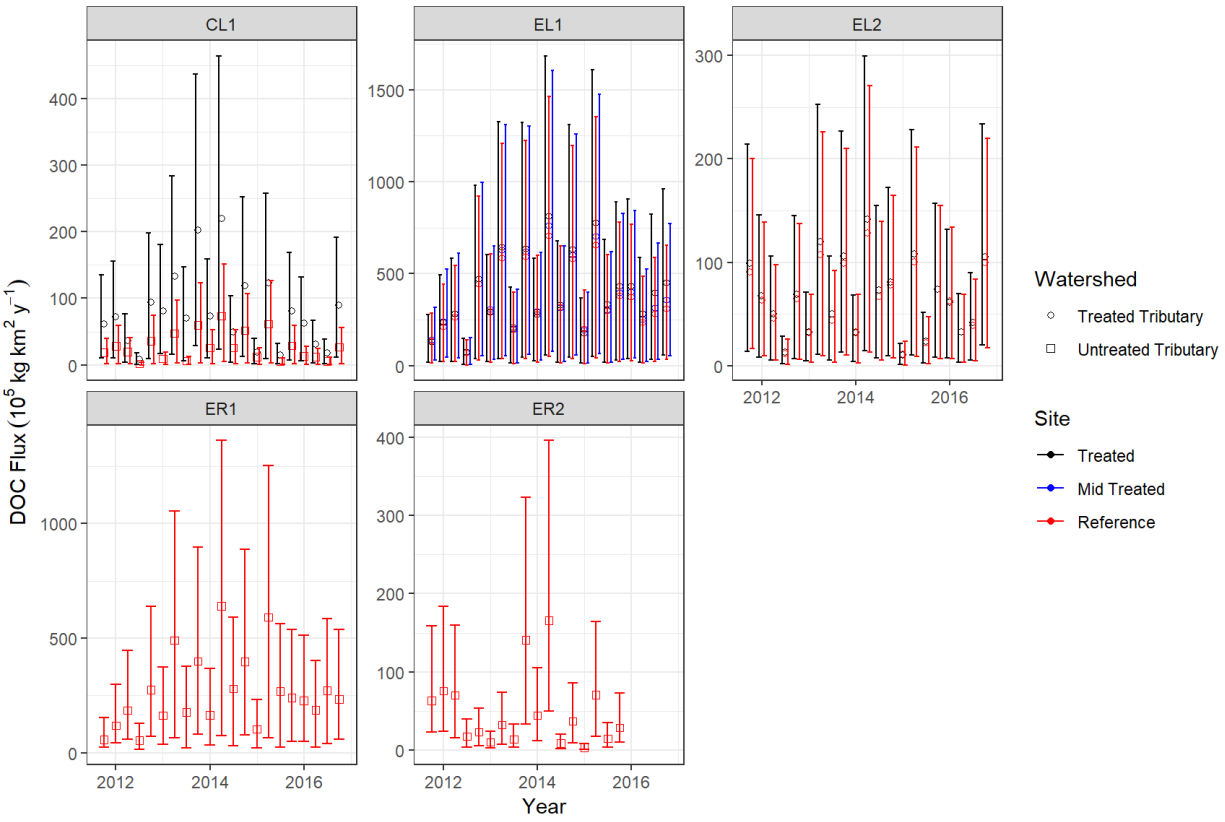

Figure 10: Seasonal loads of dissolved organic carbon for different tributaries to Honnedaga Lake. Significant differences in loads were observed only between CL1 and CR1 following calcium amendment. Channel additions did not significantly impact downstream loads.

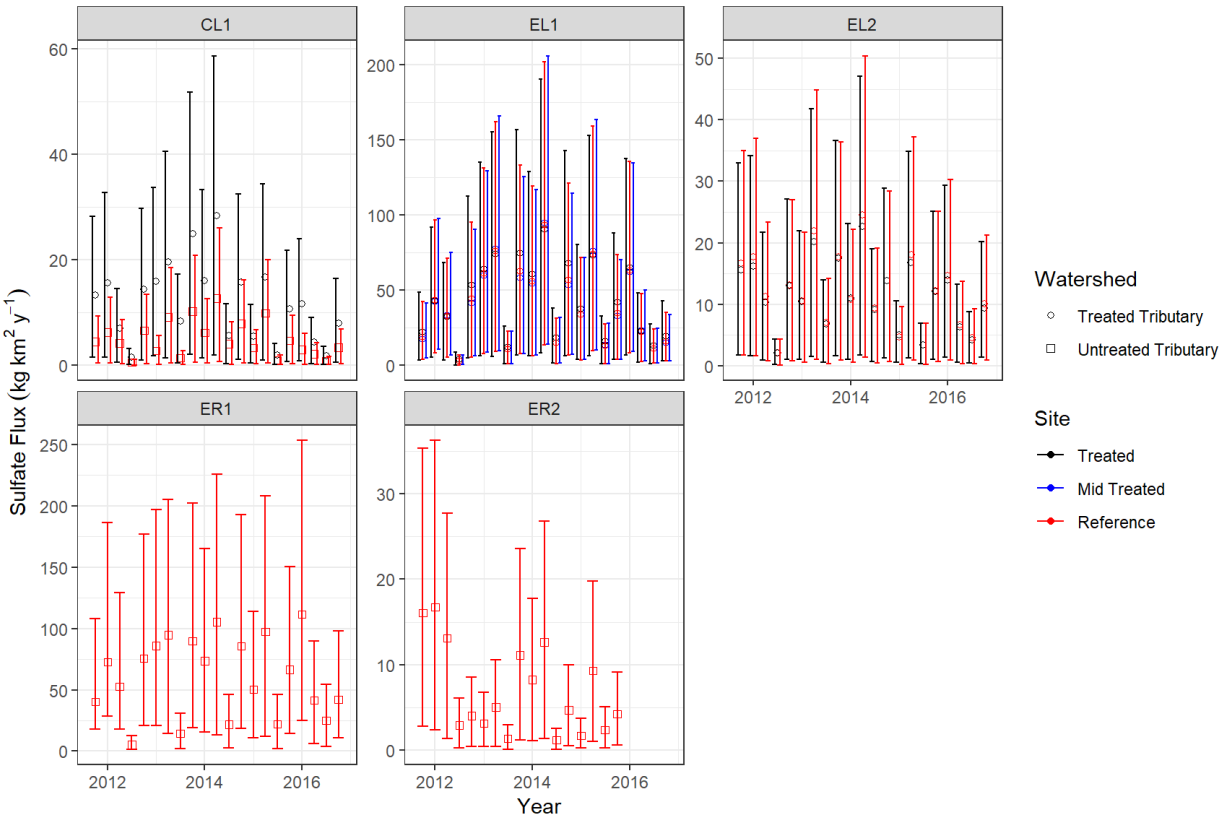

Figure 11: Seasonal loads of SO_4^{2-} for different tributaries to Honnedaga Lake. Significant differences following calcium amendment were only detected between CL1 and CR1.

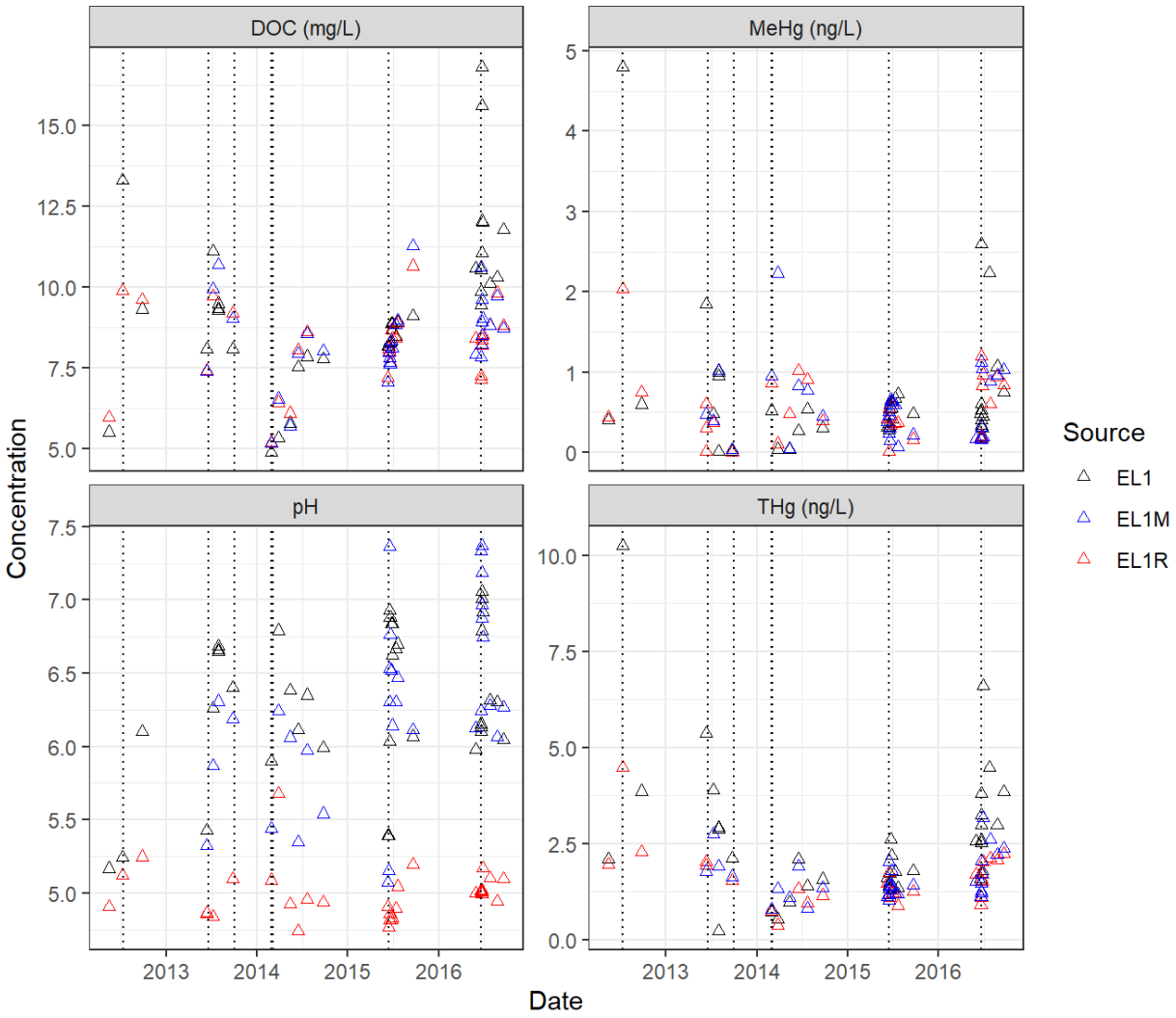

Figure 12: Chemical responses in the EL1 tributary of Honnedaga Lake to each lime addition. Intensive sampling in 2016 revealed a short-term pulse of DOC, THg, and MeHg lasting <72 hours.

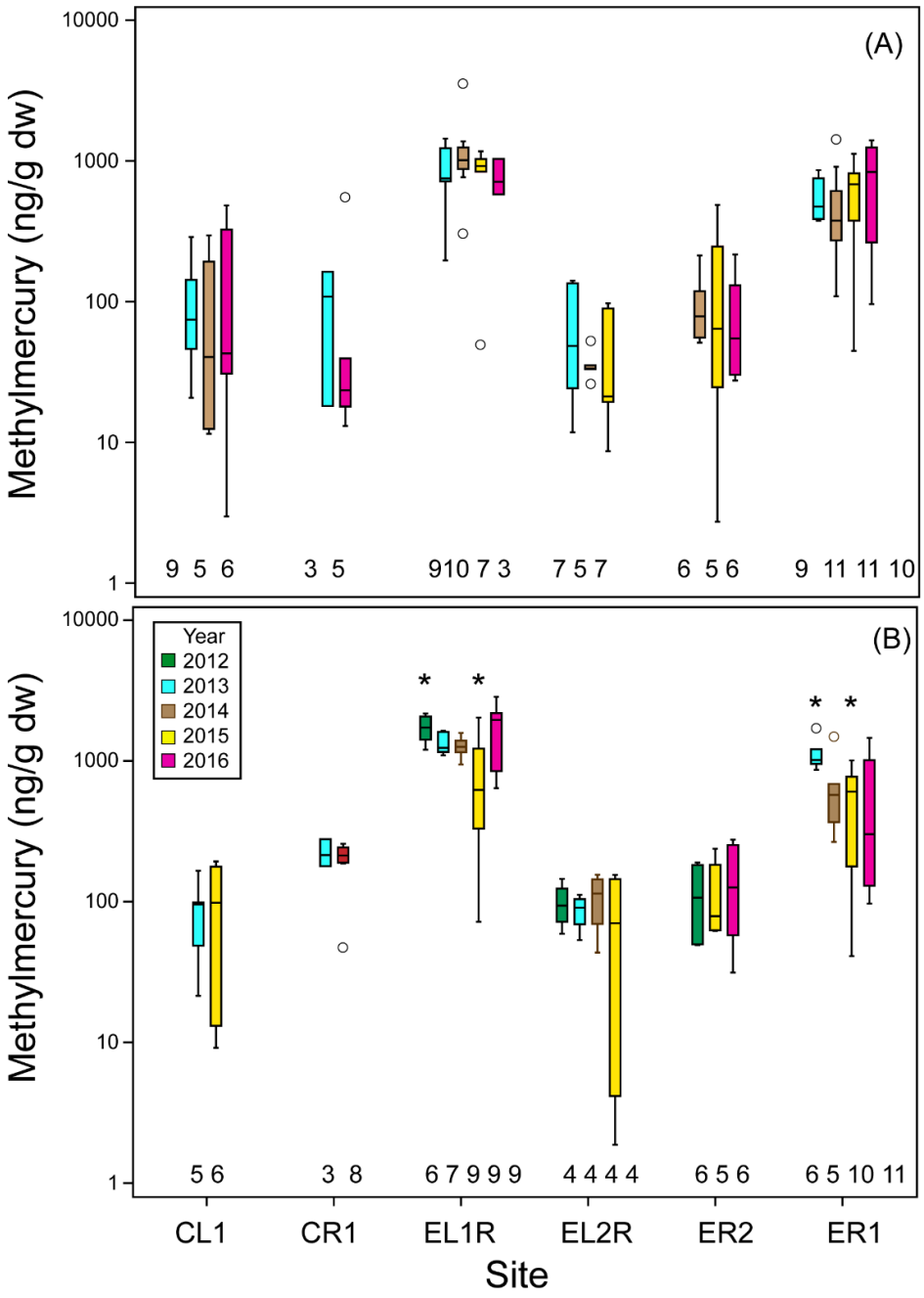

Figure 13: Comparison of annual pattern of methylmercury in macroinvertebrates from Honnedaga tributaries (normalized to trophic position 2.5) among watershed-treated (CL1) and untreated tributaries. All sites are densely-canopied except for EL1R and ER1, which have open canopies. Pre-treatment years are 2012 and 2013. Patterns are shown within seasons: (A) early summer, (B) mid-summer. Results of nonparametric analysis of variance are shown where group n >5. Numbers of samples are shown atop x axis. dw dry weight, * $p < 0.05$.

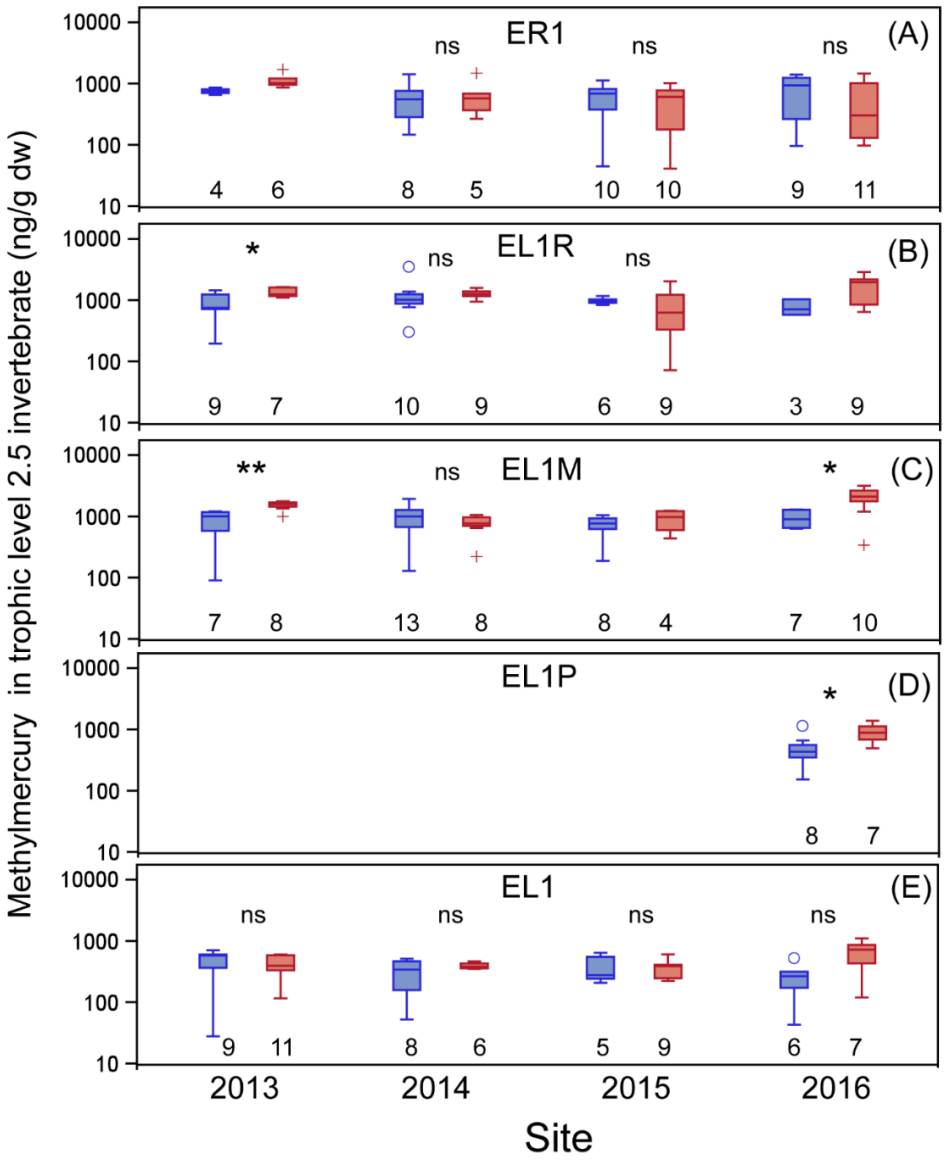

Figure 14: Methylmercury in macroinvertebrates in Honnedaga Lake tributaries normalized to trophic level 2.5 during early summer (before liming; blue boxes) and mid-summer (post liming; red boxes) in each year from 2013 -16. Seasonal and annual patterns are shown for two reference sites: (A) ER1, and (B) EL1R, and three sites located downstream of the location of lime additions that were done between the two sampling periods in 2013, 2015, and 2016 (not in 2014): (C) EL1M, located nearest to the lime addition location, (D) EL1P, located further downstream of the lime addition location yet above a large wetland complex, and EL1, located at the furthest downstream portion of the tributary and below the wetland. Data do not include northern caddisflies, which were rare in mid-summer collections. Results of rank-sum test are shown above box pairs where both groups have ≥5 samples. Numbers of samples are shown atop x axis. dw dry weight, * $p < 0.05$, ** $p < 0.01$, ns $P > 0.05$.

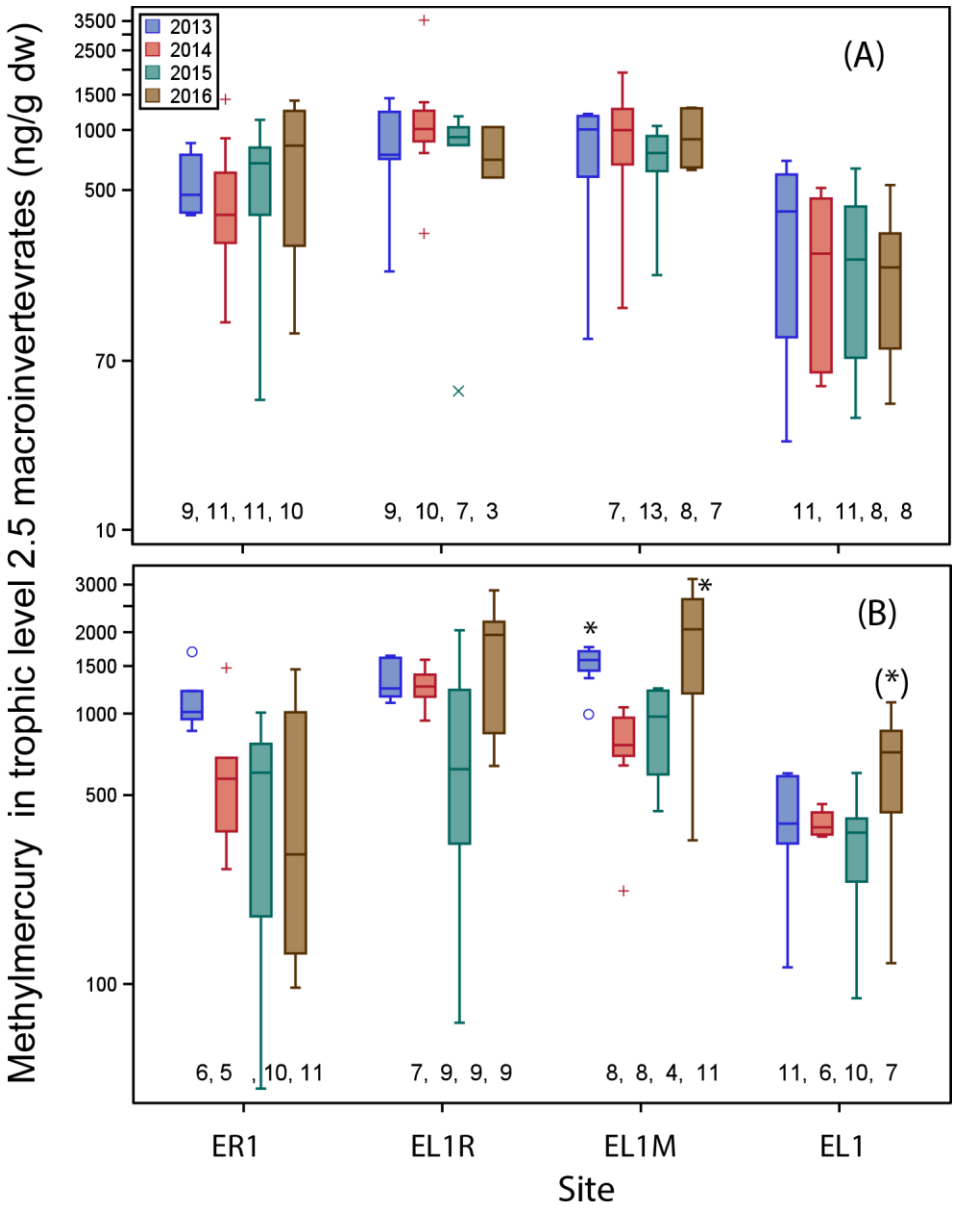

Figure 15: Comparison of macroinvertebrate methylmercury concentrations in Honnedaga Lake tributaries (normalized to trophic level 2.5) between the year 2014, when liming was done in late winter, with other years at reference (i.e. untreated) sites (ER1, EL1R) and treated sites (EL1M, EL1) in (A) early summer and (B) late summer. Asterisks above bars indicate significant difference from 2014 ($p<0.05$). Asterisk in parenthses indicates a marginally-significant difference ($p = 0.051$)

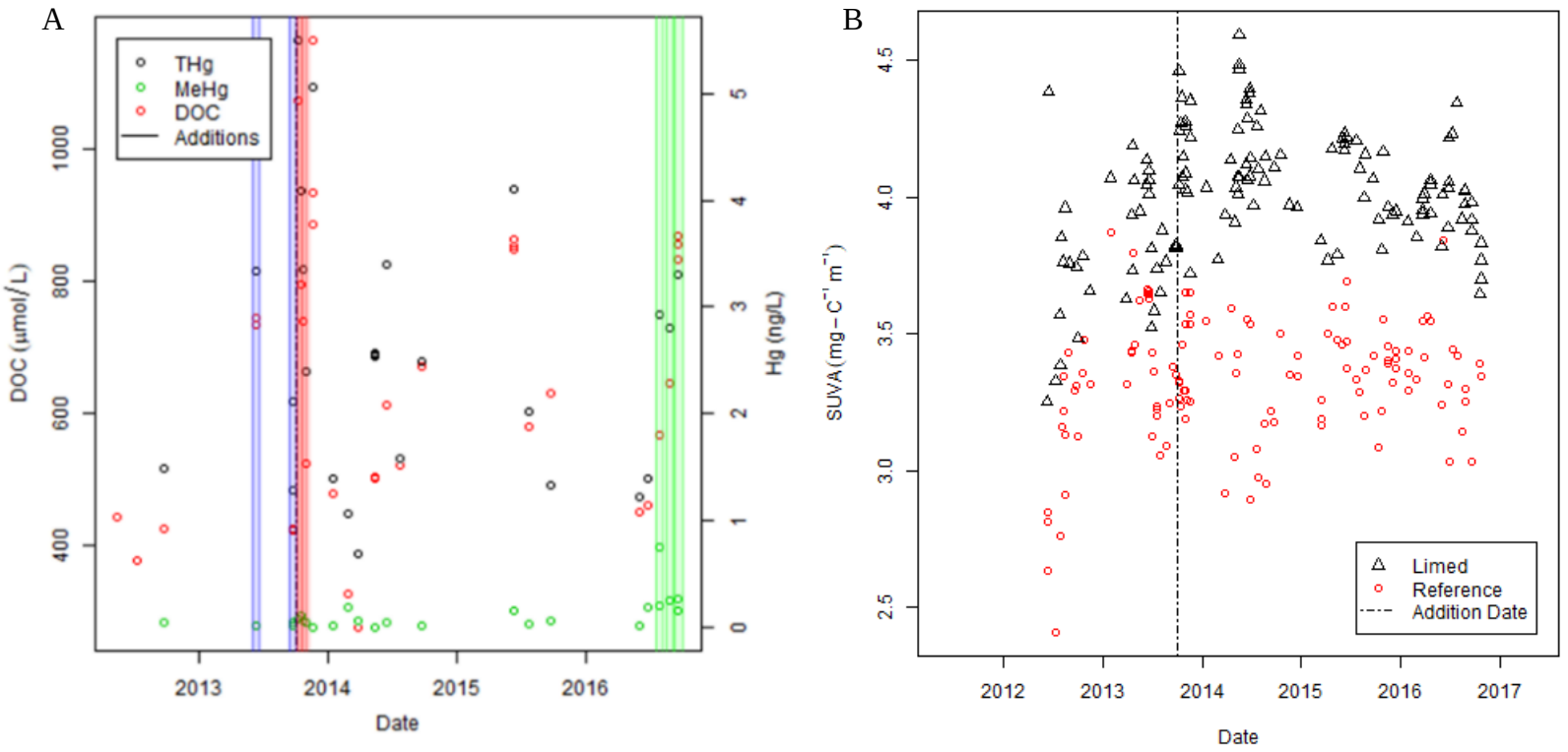

Figure 16: Time-series of DOC and mercury data from the tributary of the lime-treated watershed (A) and SUVA from both limed and reference watersheds (B) of Honnedaga Lake. Two pre-liming samples will be used to assess differences between sites before treatment. The four samples immediately following lime addition have the highest DOC and THg values with MeHg concentrations lower than the three proposed samples from 2016. The blue red and green highlighting correspond to Table 8.

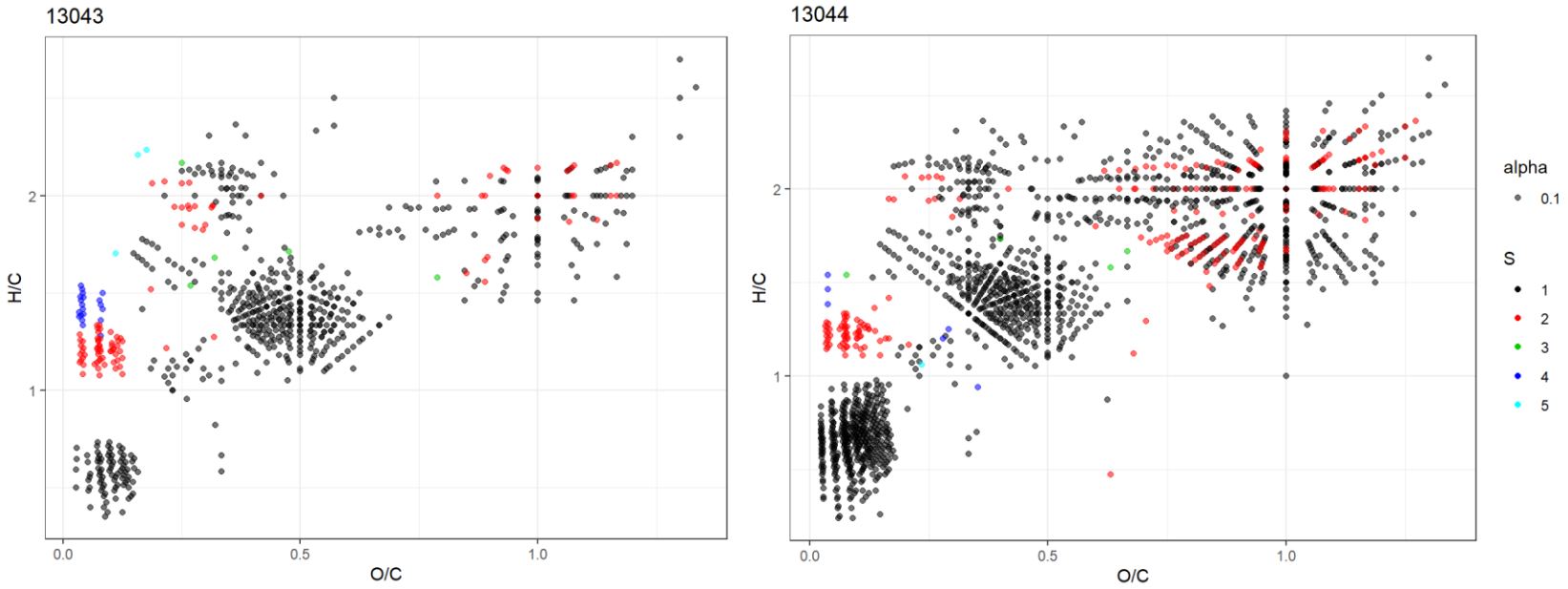

Figure 17: Van Krevelen diagrams of paired reference (13043) and treatment samples (13044) immediately following the watershed $CaCO_3$ treatment. There is an apparent shift in abundance of sulfur containing condensed and unsaturated hydrocarbons (O/C <0.21; H/C < 1.6).

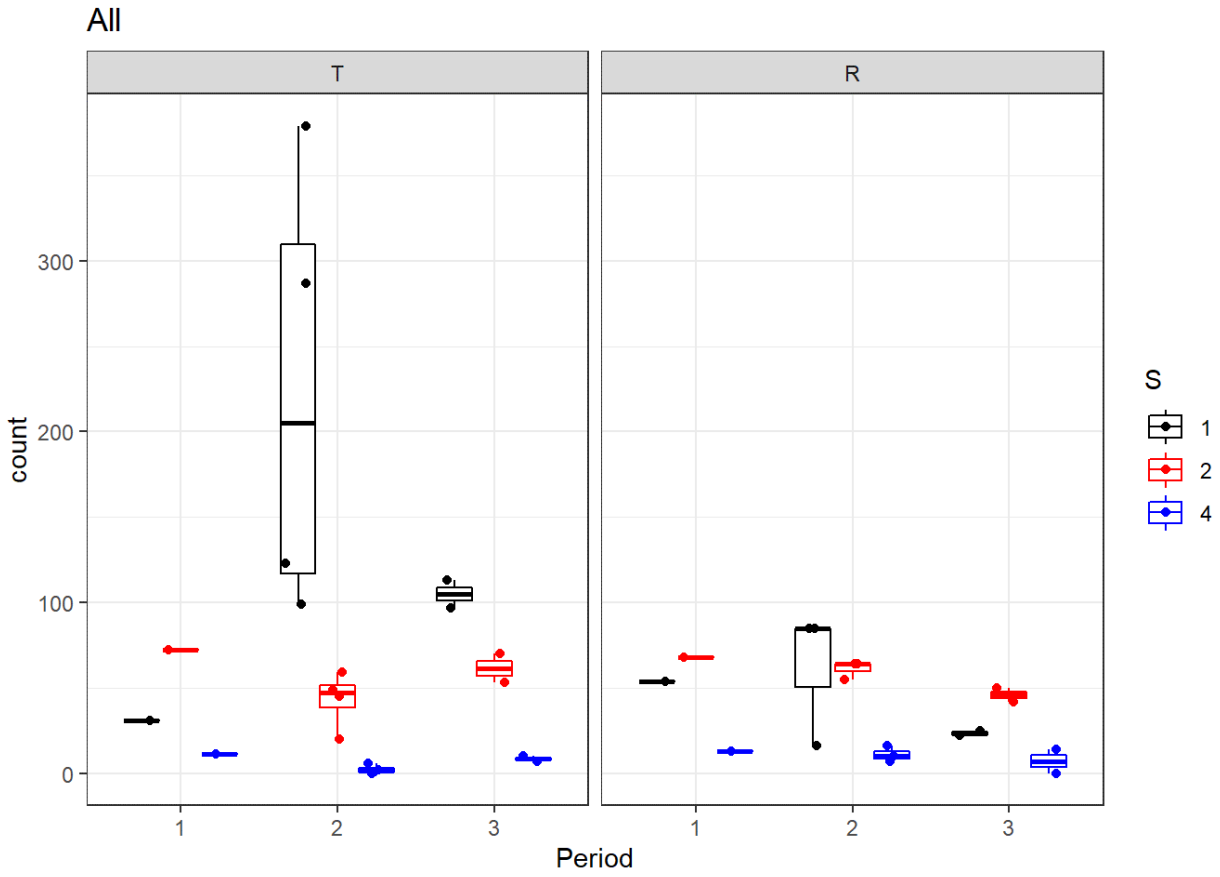

Figure 18: The relative abundance of each sulfur-containing condensed and unsaturated hydrocarbon during each treatment period at both reference and treatment sites. The legend indicates the number of sulfur atoms present in the molecular formula assignment.

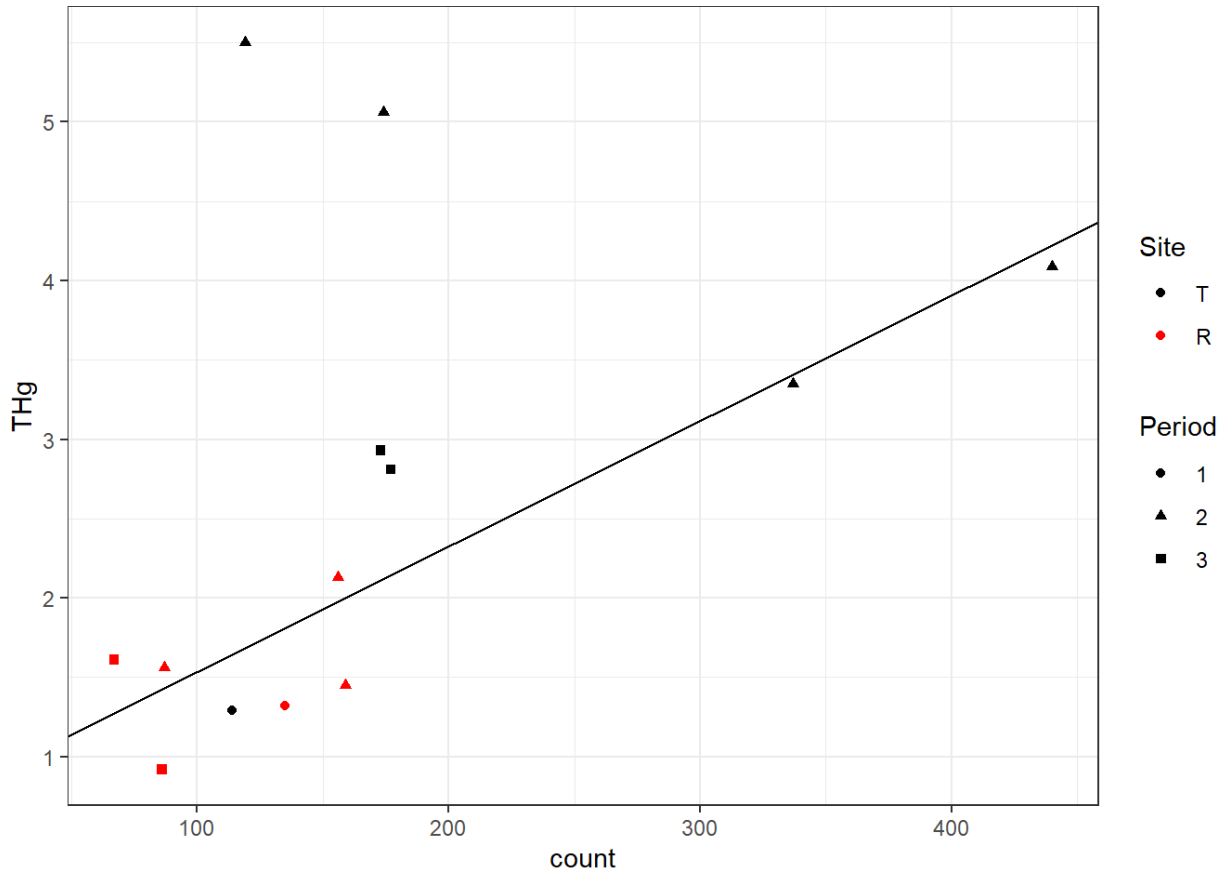

Figure 19: Linear regression across both treatment and reference samples, excluding the two treatment outliers immediately following $CaCO_3$ application ($m = 0.008$, $p < 0.001$). T = treatment; R = reference.

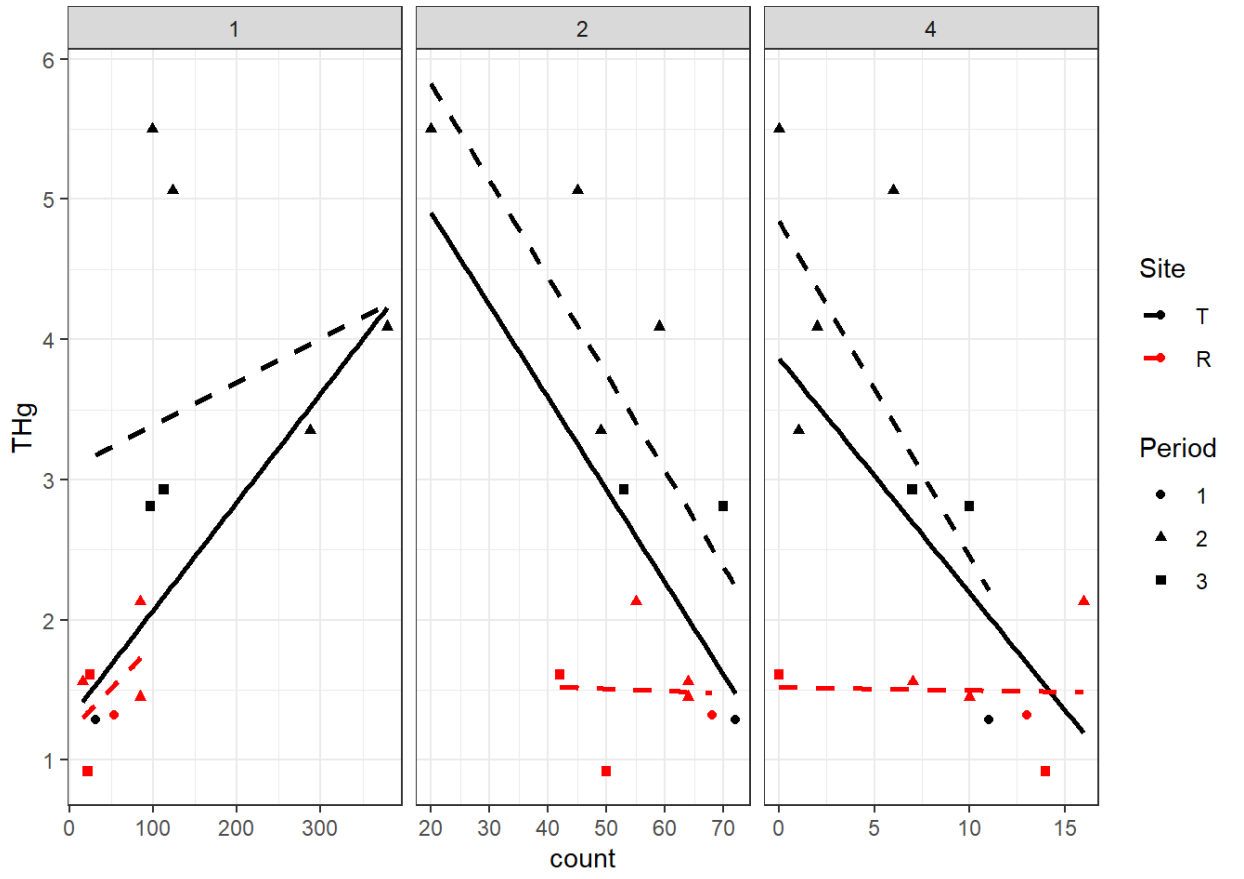

Figure 20: Linear regressions between THg and the total number of monosulfur condensed and unsaturated hydrocarbons. Solid lines are across both treatment and reference sites and hashed lines are for a single site. T= treatment; R=reference

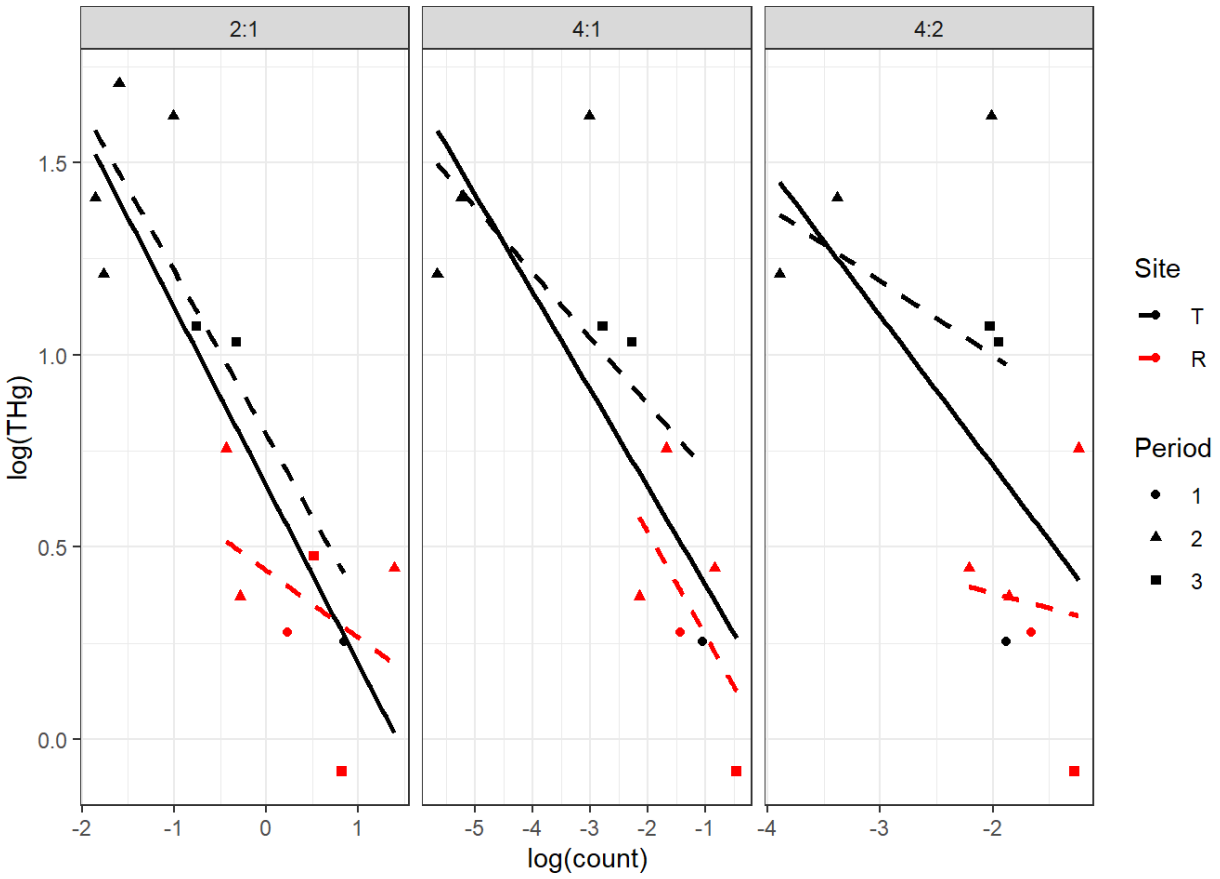

Figure 21: Linear regression between the natural logarithm of THg and the ratio between sulfur-containing hydrocarbons. T= treatment, R=reference.

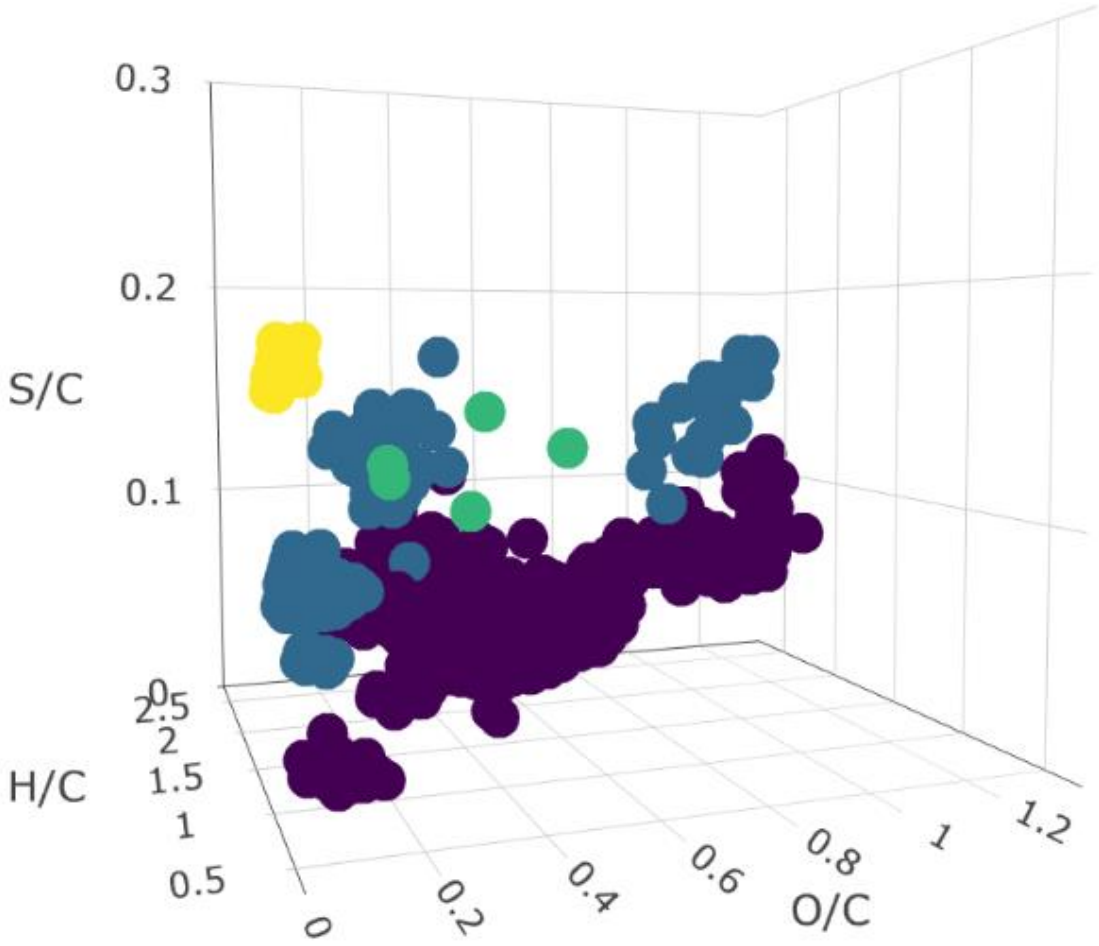

Figure 22: 3-D Van Krevelen diagram of the sulfur-containing assignments in the pre-treatment sample. All samples presented similar distributions of data. A lighter color indicates a higher number of sulfur (yellow = 4, green = 3, blue =2, violet = 1). The number of assignments in the most carbon dense region had a significant positive relationship with THg. O/C = oxygen to carbon ratio; H/C = hydrogen to carbon ratio, S/C = sulfur to carbon ratio.

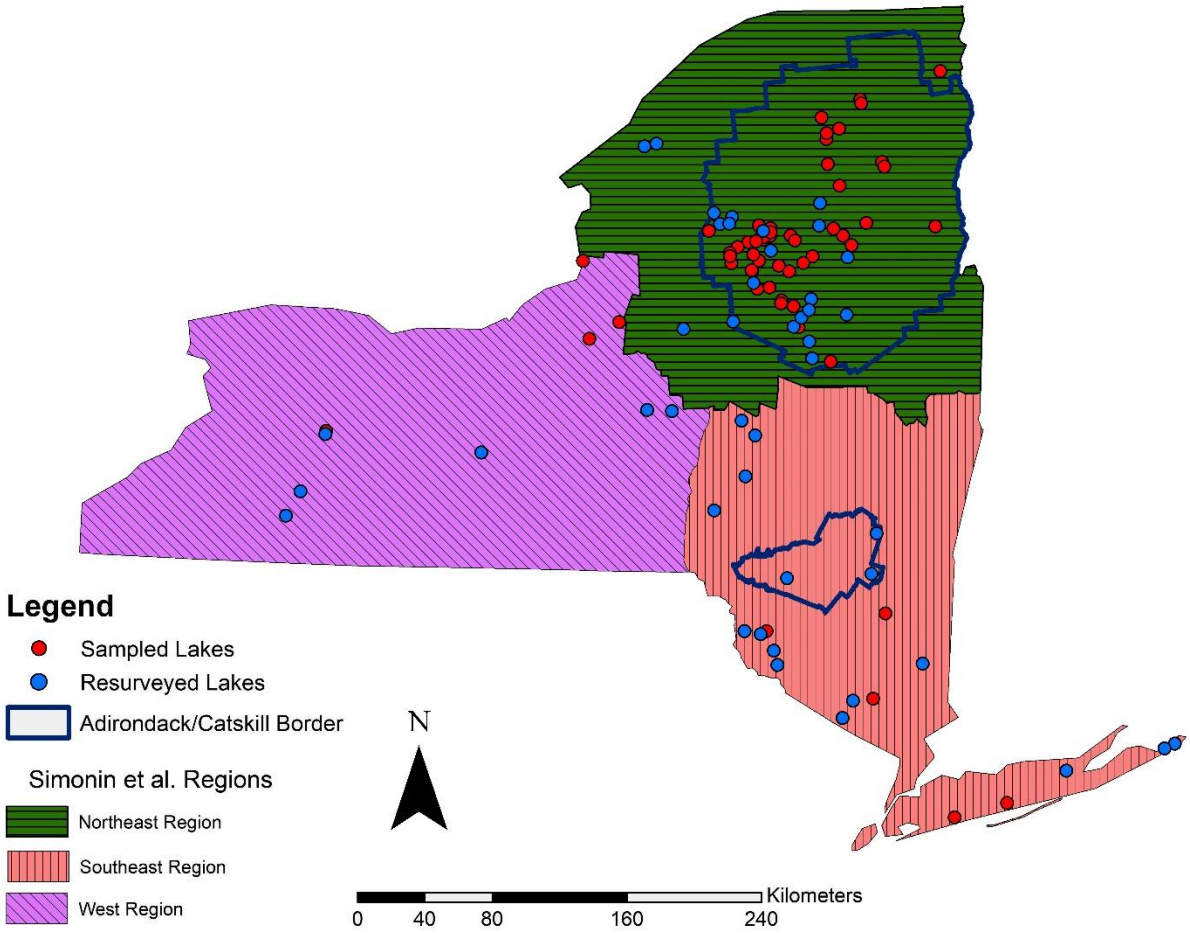

Figure 23: Lakes sampled as part of the 2010s resurvey. Lakes that were also sampled as part of the 2000s survey are in blue. The three regions of New York State were originally used by Simonin et al. 2008.

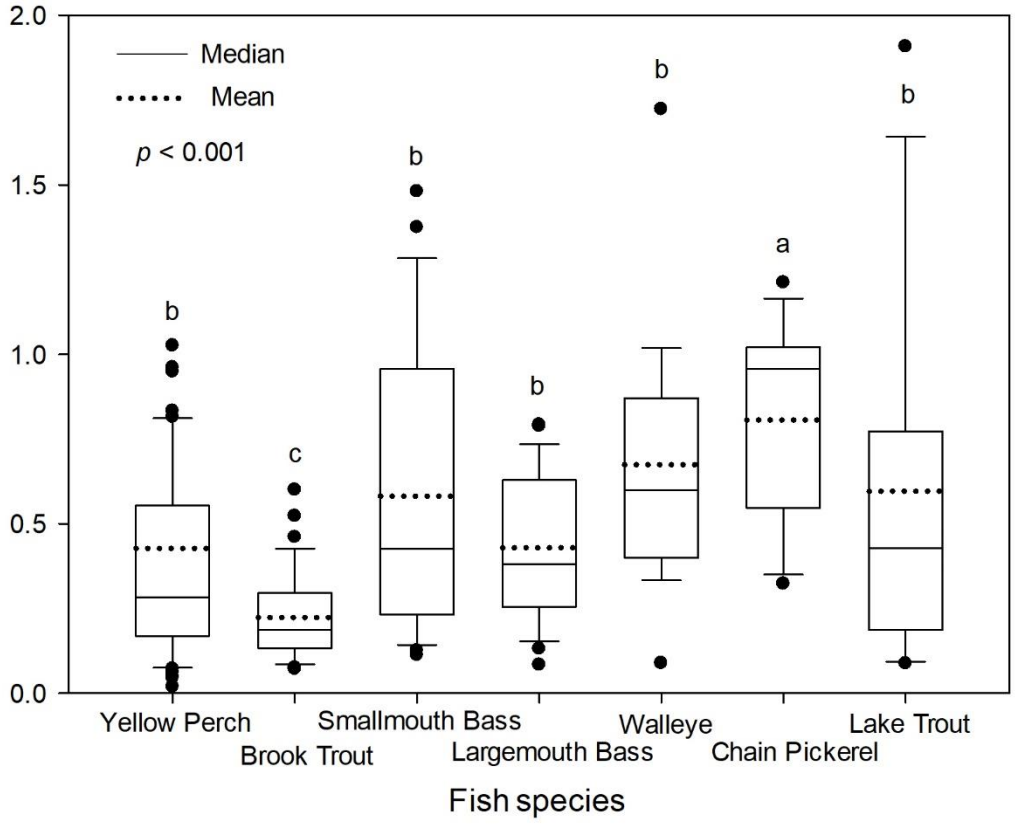

Figure 24: Standard-size THg concentrations in seven fish species from 2014-2016 in New York State lakes. Bars with different letters indicated the concentrations were different among species.

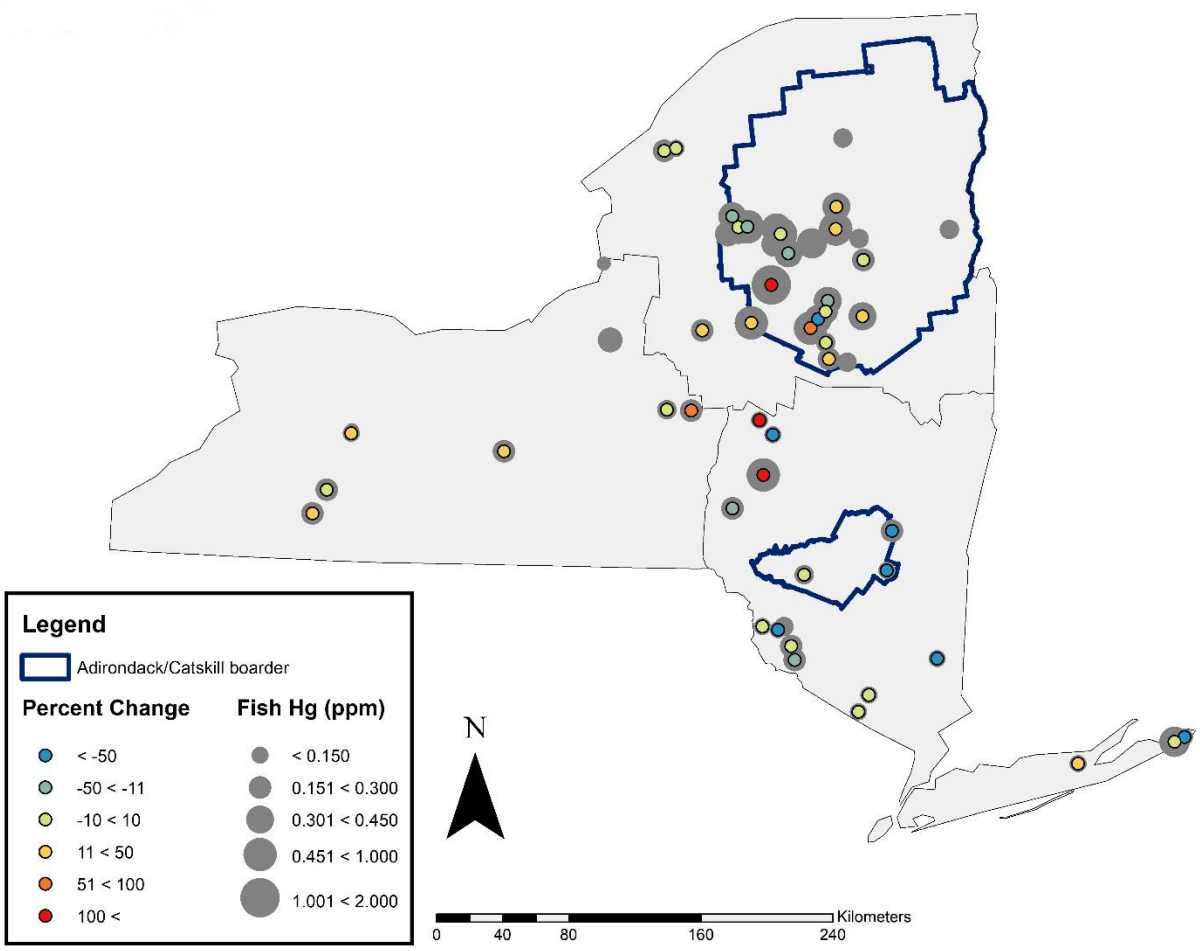

Figure 25: Yellow perch concentrations were highest in the Northeast region of NYS. Fish Hg increased between 2000s and 2010s surveys in both the Northeast and West regions with modest reductions in the Southeast.

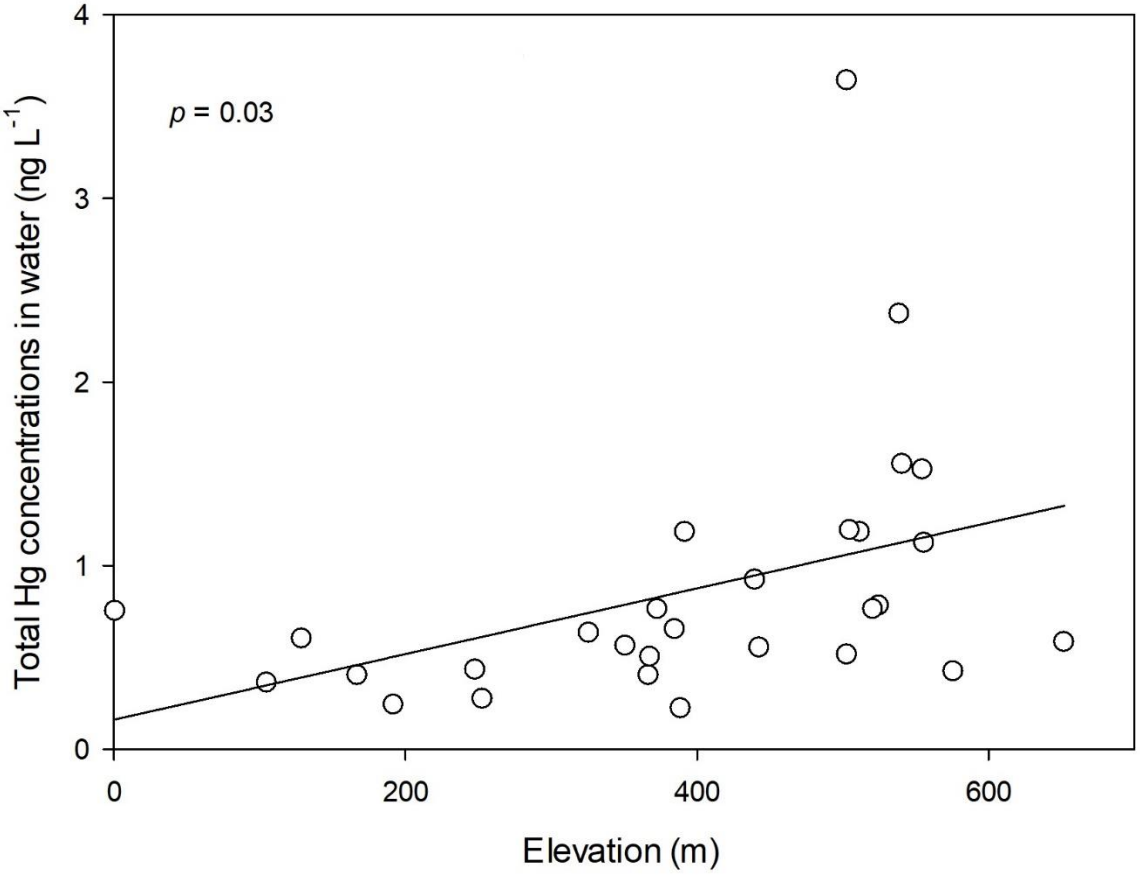

Figure 26: THg concentrations in surface water with lake elevation.

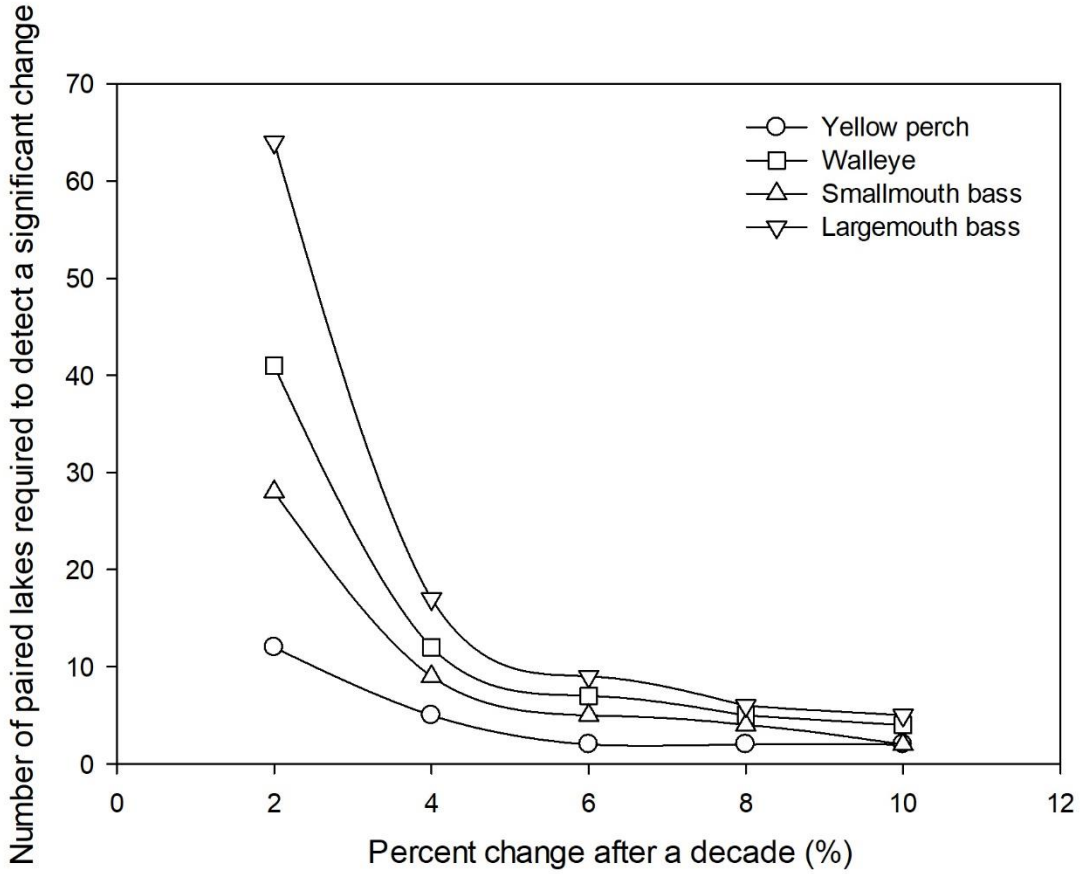

Figure 27: Number of lakes required to detect a change in fish THg concentrations for four common species after a decade in New York State.

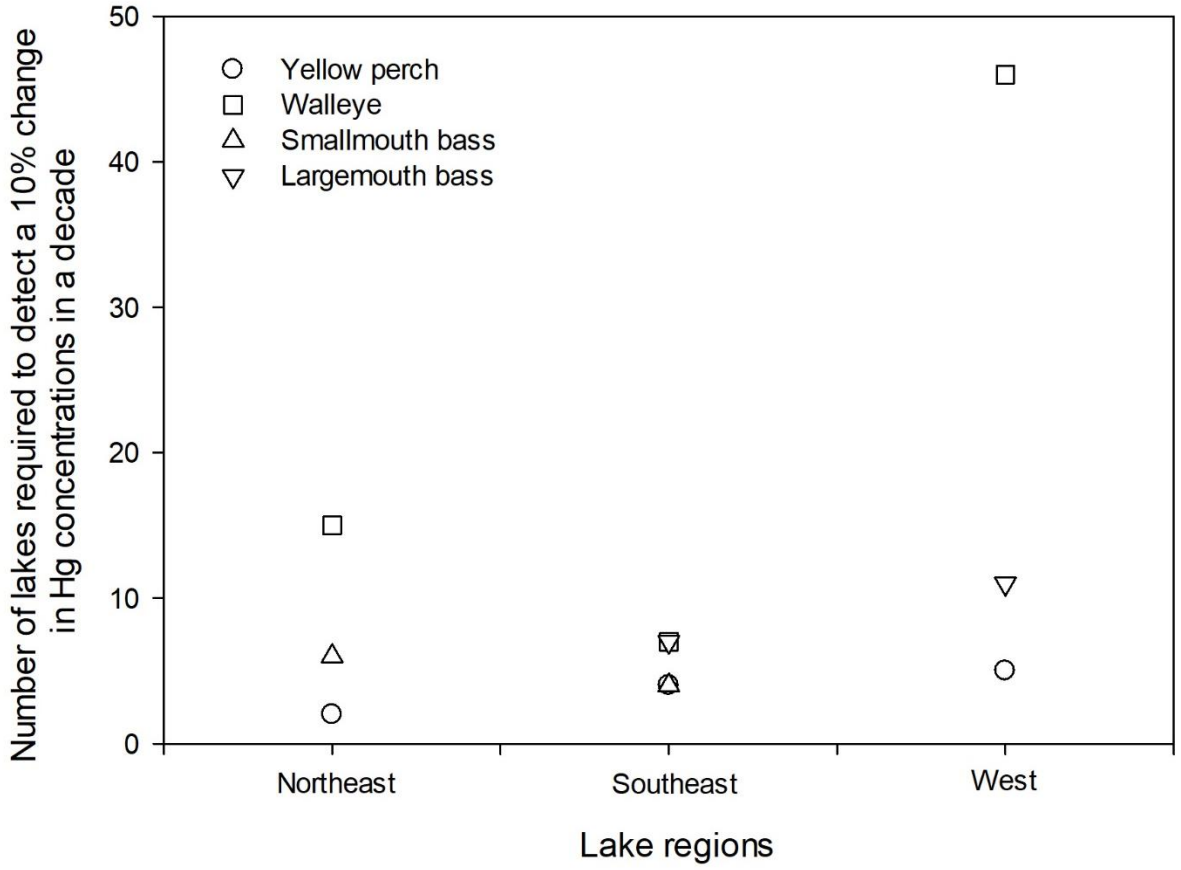

Figure 28: Number of lakes required to detect a 10% change in fish THg concentrations in four common species after a decade for different regions in New York State.

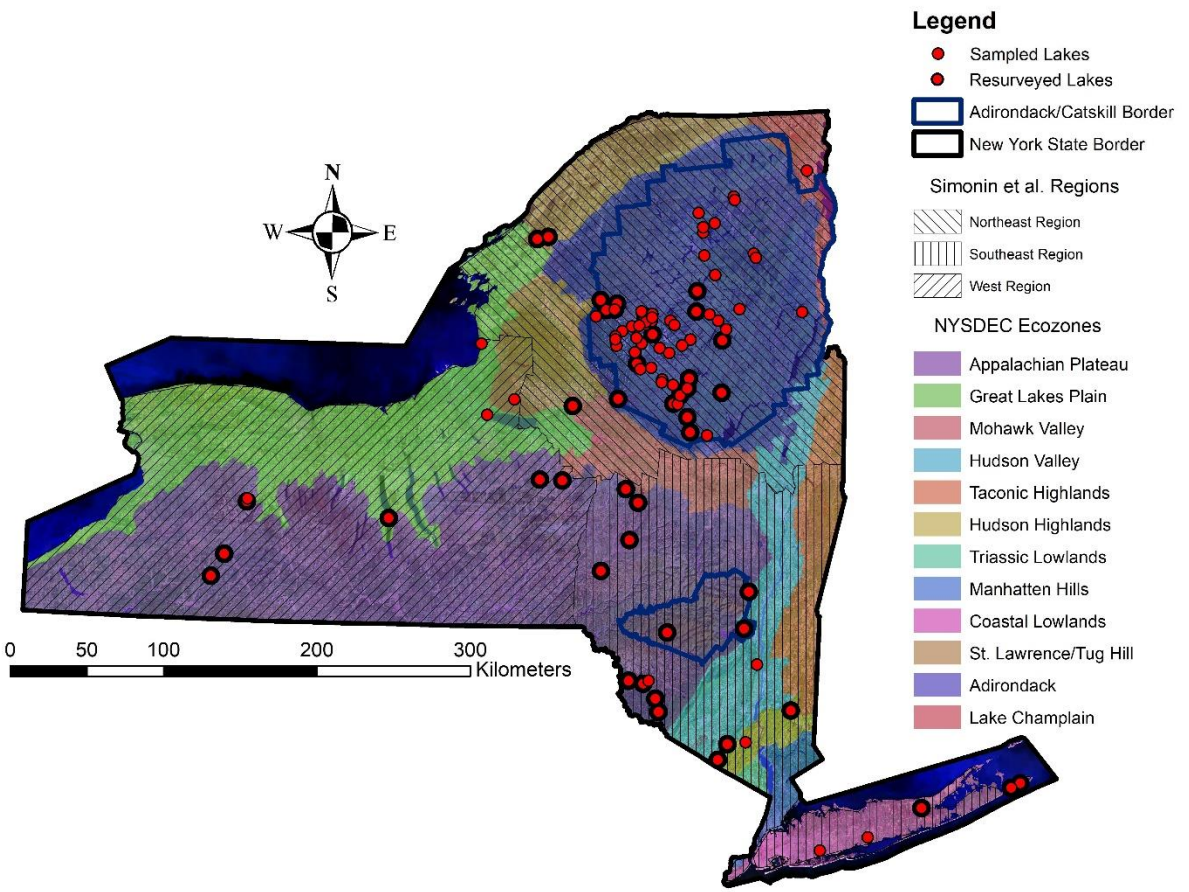

Figure 29: Samples collected as part of the 2010 survey, with 2000 outlined in bold. There is coverage of almost all 12 ecoregions as defined by the NYSDEC.

9. Tables

Table 1: Site names, acidification and treatment type, location information, and USGS station identifiers.

site code	Site name	USGS station identifier	Latitude	Longitude	site type
ER1	Middle Branch Black River near Forest Lodge NY	425078820	43.5397222	-74.8364444	untreated tributary
CL1	Honnedaga Lake Tributary No 3 near Forest Lodge NY	0134277114	43.53138889	-74.8529167	chronically-acidic, treated watershed
CR1	Honnedaga Lake Tributary No 4 North of Forest Lodg	0134277112	43.53380556	-74.8618333	untreated; reference site for watershed-treatment site
EL1R	Honnedaga Lake Tributary No 4 North of Forest Lodg	0134277118	43.5302222	-74.80269444	untreated location upstream of in-channel liming location
EL1	Honnedaga Lake Tributary No 4 at Forest Lodge NY	0134277119	43.52105556	-74.8011111	episodically-acidic; treated; downstream of in-channel lime application site
EL1M	Honnedaga Lake Tributary No 4 near Forest Lodge NY	013427711805	43.5283333	-74.8019444	episodically-acidic; treated; downstream of in-channel lime application site
EL1P	Honnedaga Lake Tributary No 4 Upstream of Forest L	01342771835	43.52644444	-74.8014444	episodically-acidic; treated; downstream of in-channel lime application site
EL2R	Honnedaga Lake Tributary No 5 Southeast of Forest	0134277120	43.5135833	-74.7979722	untreated location upstream of in-channel liming location
EL2	Honnedaga Lake Trib No 5 at Forest Lodge NY	0134277121	43.51430556	-74.7988611	episodically-acidic; treated; downstream of in-channel lime application site
ER2	Honnedaga Lake Tributary No 6 at Forest Lodge NY	0134277123	43.5115	-74.8364444	untreated tributary

Table 2: Macroinvertebrate sample field and laboratory data. Site names corresponding with site codes are provided in Table S1. ES early summer, LS late summer, SP spring, FC filtering collector, GC gathering collector, PR predator, SC scraper, SH shredder.

site code	season	feeding group	Common name	Scientific name	feeding group - taxon code	Sample collection date	N15:N14 (per mil)	C13:C12 (per mil)	Individuals in sample	Methylmercury (ng/g dry weight)	Field weight of sample (g)	N15:14 of base consumer for site (per mil)	Calculated methylmercury in trophic position 2.5 consumer (ng/g dry weight)
CL1	ES	FC	Net-spinning caddisfly	Hydropsychidae	FC2	6/11/2013	5.8	-24.4	9	97.09	0.2	1.06	101.39
CL1	ES	GC	Crayfish	Cambaridae	GC2	6/11/2013	3.7	-24.9	2	32.77	2.2	1.06	46.12
CL1	ES	GC	Crayfish	Cambaridae	GC2	6/11/2013	2.7	-25.5	2	130.62	2.3	1.06	220.28
CL1	ES	GC	Crayfish	Cambaridae	GC2	6/11/2013	2.6	-24.5	1	167.15	1.5	1.06	287.61
CL1	ES	PR	Crane fly	Tipulidae	PR1	6/11/2013	7.1	-24.1	3	82.69	0.7	1.06	74.46
CL1	ES	PR	Darner dragonfly	Aeshnidae	PR1	6/11/2013	4.6	-27.1	2	116.63	0.4	1.06	142.84
CL1	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/11/2013	0.5	-25.4	4	6.92	0.5	1.06	20.70
CL1	ES	SH	Rolled-winged stoneflies	Leuctridae	SH2	6/11/2013	3.8	-25.5	109	35.36	0.4	1.06	48.95
CL1	ES	SH	Shredding stonefly	Plecoptera	SH2	6/11/2013	1.3	-27.2	105	13.16	0.2	1.06	30.72
CL1	LS	GC	Crayfish	Cambaridae	GC2	7/31/2013	2	-25.3	1	84.54	1.2	1.06	165.58
CL1	LS	PR	Darner dragonfly	Aeshnidae	PR1	7/31/2013	4.7	-26	1	79.28	0.4	1.06	95.72
CL1	LS	SH	Northern caddisfly	Limnephilidae	SH1	7/31/2013	1.4	-26.4	111	43.52	0.3	1.06	98.91
CL1	LS	SH	Northern caddisfly	Limnephilidae	SH1	7/31/2013	1.4	-25.5	3	9.42	0.3	1.06	21.40
CL1	LS	SH	Shredding stonefly	Plecoptera	SH2	7/31/2013	3.6	-26	116	33.96	0.3	1.06	48.60
CL1	SP	GC	Crayfish	Cambaridae	GC2	5/13/2014	3.17	-23.95	1	99.73	0.57	1.06	153.85
CL1	SP	PR	Common stonefly	Perlidae	PR1	5/13/2014	7.29	-25.66	4	24.36	0.146	1.06	21.50

site code	season	feeding group	Common name	Scientific name	feeding group - taxon code	Sample collection date	N15:N14 (per mil)	C13:C12 (per mil)	Individuals in sample	Methylmercury (ng/g dry weight)	Field weight of sample (g)	N15:14 of base consumer for site (per mil)	Calculated methylmercury in trophic position 2.5 consumer (ng/g dry weight)
CL1	SP	SH	Northern caddisfly	Limnephilidae	SH1	5/13/2014	1.82	-26.53	6	39.53	0.508	1.06	80.76
CL1	SP	SH	Northern caddisfly	Limnephilidae	SH1	5/13/2014	1.63	-26.24	5	4.20	0.462	1.06	8.99
CL1	SP	SH	Northern caddisfly	Limnephilidae	SH1	5/13/2014	1.63	-26.12	5	31.65	0.454	1.06	67.76
CL1	SP	SH	Rolled-winged stoneflies	Leuctridae	SH2	5/13/2014	5.15	-25.1	54	45.73	0.09	1.06	51.90
CL1	ES	GC	Crayfish	Cambaridae	GC2	6/17/2014	2.33	-25.29	1	6.32	7.68	1.06	11.51
CL1	ES	GC	Crayfish	Cambaridae	GC2	6/17/2014	2.54	-26.8	1	110.57	4.22	1.06	192.60
CL1	ES	GC	Crayfish	Cambaridae	GC2	6/17/2014	2.52	-25.42	1	168.86	1.88	1.06	295.33
CL1	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/17/2014	0.62	-26.44	5	4.33	0.38	1.06	12.45
CL1	ES	SH	Shredding stonefly	Plecoptera	SH2	6/17/2014	2.81	-29.5	85	24.48	0.28	1.06	40.40
CL1	ES	GC	Crayfish	Cambaridae	GC2	6/10/2015	2.07	-25.68	1	142.26	2.14	1.06	274.21
CL1	ES	GC	Crayfish	Cambaridae	GC2	6/10/2015	3.02	-27.22	1	---	4.84	1.06	---
CL1	ES	PR	Alderfly	Sialidae	PR1	6/10/2015	6.08	-25.38	5	---	0.08	1.06	---
CL1	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/10/2015	1.09	-25.89	6	114.59	0.57	1.06	283.98
CL1	ES	SH	Shredding stonefly	Plecoptera	SH2	6/10/2015	3.53	-26.5	7	---	0.01	1.06	---
CL1	LS	GC	Crayfish	Cambaridae	GC2	7/22/2015	1.26	-24.95	6	63.64	0.57	1.06	150.27
CL1	LS	GC	Crayfish	Cambaridae	GC2	7/22/2015	1.68	-24.86	1	91.25	2.14	1.06	192.94
CL1	LS	GC	Crayfish	Cambaridae	GC2	7/22/2015	1.8	-25.65	1	4.46	4.84	1.06	9.15
CL1	LS	GC	Crayfish	Cambaridae	GC2	7/22/2015	1.35	-24.83		76.92		1.06	177.18
CL1	LS	SH	Shredding stonefly	Plecoptera	SH2	7/22/2015	4.28	-25.5	122	10.22	0.27	1.06	13.12

site code	season	feeding group	Common name	Scientific name	feeding group - taxon code	Sample collection date	N15:N14 (per mil)	C13:C12 (per mil)	Individuals in sample	Methylmercury (ng/g dry weight)	Field weight of sample (g)	N15:14 of base consumer for site (per mil)	Calculated methylmercury in trophic position 2.5 consumer (ng/g dry weight)
CL1	LS	SH	Shredding stonefly	Plecoptera	SH2	7/22/2015	4.3	-25.46	122	35.97	0.25	1.06	46.05
CL1	ES	GC	Crayfish	Cambaridae	GC2	5/31/2016	2.55	-25.78	1	277.67	5.39	1.06	482.66
CL1	ES	GC	Crayfish	Cambaridae	GC2	5/31/2016	2	-26.29	1	165.81	0.76	1.06	324.75
CL1	ES	PR	Alderfly	Sialidae	PR1	5/31/2016	5.1	-24.93	3	---	0.05	1.06	---
CL1	ES	SH	Northern caddisfly	Limnephilidae	SH1	5/31/2016	0.43	-26.02	4	10.02	0.36	1.06	30.74
CL1	ES	SH	Northern caddisfly	Limnephilidae	SH1	5/31/2016	0.51	-26.06	4	1.00	0.47	1.06	2.97
CL1	ES	SH	Northern caddisfly	Limnephilidae	SH1	5/31/2016	0.62	-26.63	4	11.88	0.45	1.06	34.11
CL1	ES	SH	Shredding stonefly	Plecoptera	SH2	5/31/2016	3.23	-26.92	86	33.83	0.2	1.06	51.63
CR1	ES	GC	Crayfish	Cambaridae	GC2	6/11/2013	2.5	-25.9	1	99.30	1.6	0.72	162.95
CR1	ES	PR	Crane fly	Tipulidae	PR1	6/11/2013	4.4	-24.4	2	90.24	0.4	0.72	108.33
CR1	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/11/2013	0.1	-25.2	6	5.92	0.3	0.72	18.11
CR1	LS	FC	Net-spinning caddisfly	Hydropsychidae	FC2	7/31/2013	4.3	-26.3	25	146.76	0.1	0.72	178.71
CR1	LS	GC	Crayfish	Cambaridae	GC2	7/31/2013	2.2	-26.7	1	123.05	1.6	0.72	214.32
CR1	LS	GC	Crayfish	Cambaridae	GC2	7/31/2013	2.1	-24.5	1	156.45	3.1	0.72	278.20
CR1	LS	SH	Northern caddisfly	Limnephilidae	SH1	7/31/2013	1.4	-26.3	72	---	0.4	0.72	---
CR1	SP	FC	Net-spinning caddisfly	Hydropsychidae	FC2	5/13/2014	5.05	-25.63	13	125.65	0.208	0.72	138.17
CR1	SP	PR	Crane fly	Tipulidae	PR1	5/13/2014	6.83	-24.42	2	81.61	0.5	0.72	72.94
CR1	SP	PR	Crane fly	Tipulidae	PR1	5/13/2014	6.14	-23.72	5	83.59	0.06	0.72	80.56
CR1	SP	SH	Northern caddisfly	Limnephilidae	SH1	5/13/2014	1.88	-27.41	5	30.55	0.278	0.72	56.95

site code	season	feeding group	Common name	Scientific name	feeding group - taxon code	Sample collection date	N15:N14 (per mil)	C13:C12 (per mil)	Individuals in sample	Methylmercury (ng/g dry weight)	Field weight of sample (g)	N15:14 of base consumer for site (per mil)	Calculated methylmercury in trophic position 2.5 consumer (ng/g dry weight)
CR1	SP	SH	Northern caddisfly	Limnephilidae	SH1	5/13/2014	2.11	-27.18	5	3.90	0.39	0.72	6.93
CR1	SP	SH	Northern caddisfly	Limnephilidae	SH1	5/13/2014	3.53	-25.86	4	2.04	0.258	0.72	2.79
CR1	SP	SH	Shredding stonefly	Plecoptera	SH2	5/13/2014	3.09	-26.31	63	32.18	0.126	0.72	47.40
CR1	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/17/2014	0.36	-26.7	2	12.62	0.11	0.72	35.28
CR1	ES	SH	Shredding stonefly	Plecoptera	SH2	6/17/2014	3.69	-25.72	37	46.21	0.07	0.72	61.67
CR1	LS	SH	Northern caddisfly	Limnephilidae	SH1	7/22/2014	0.6	-26.43	58	28.19	0.12	0.72	73.06
CR1	ES	PR	Crane fly	Tipula	PR1	6/10/2015	5.68	-24.54	1	---	0.22	0.72	---
CR1	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/10/2015	0.59	-26.2	11	14.64	0.44	0.72	38.06
CR1	ES	SH	Shredding stonefly	Plecoptera	SH2	6/10/2015	3.31	-26.08	86	---	0.18	0.72	---
CR1	LS	FC	Net-spinning caddisfly	Hydropsychidae	FC2	7/22/2015	2.45	-25.98	10	---	0.04	0.72	---
CR1	LS	GC	Crayfish	Cambaridae	GC2	7/22/2015	1.91	-26.07	1	123.21	6.75	0.72	228.17
CR1	LS	GC	Crayfish	Cambaridae	GC2	7/22/2015	2.03	-25.46	1	142.67	4.07	0.72	257.46
CR1	LS	GC	Crayfish	Cambaridae	GC2	7/22/2015	1.82	-25.93	1	112.88	3.15	0.72	213.22
CR1	LS	GC	Crayfish	Cambaridae	GC2	7/22/2015	2.29	-25.02	1	124.52	3.03	0.72	212.96
CR1	LS	GC	Crayfish	Cambaridae	GC2	7/22/2015	2.82	-25.69	1	166.95	1.2	0.72	258.01
CR1	LS	GC	Crayfish	Cambaridae	GC2	7/22/2015	2.09	-25.62	1	107.62	1.46	0.72	191.77
CR1	LS	GC	Crayfish	Cambaridae	GC2	7/22/2015	5.36	-25.54	1	176.47	1.09	0.72	186.57
CR1	LS	PR	Alderfly	Sialidae	PR1	7/22/2015	5.4	-24.31	1	---	0.01	0.72	---
CR1	LS	PR	Predaceous diving beetle	Dytiscidae	PR1	7/22/2015	3.12	-26.29	2	---	0.03	0.72	---

site code	season	feeding group	Common name	Scientific name	feeding group - taxon code	Sample collection date	N15:N14 (per mil)	C13:C12 (per mil)	Individuals in sample	Methylmercury (ng/g dry weight)	Field weight of sample (g)	N15:14 of base consumer for site (per mil)	Calculated methylmercury in trophic position 2.5 consumer (ng/g dry weight)
CR1	LS	SH	Caddisfly	Trichoptera	SH1	7/22/2015	-1.59	-26.7	14	---	0.06	0.72	---
CR1	LS	SH	Shredding stonefly	Plecoptera	SH2	7/22/2015	2.98	-26.14	108	31.22	0.17	0.72	46.89
CR1	ES	GC	Crayfish	Cambaridae	GC2	5/31/2016	4.35	-26.85	1	455.83	6.04	0.72	551.14
CR1	ES	SH	Northern caddisfly	Limnephilidae	SH1	5/31/2016	-0.3	-25.16	18	5.03	0.52	0.72	17.95
CR1	ES	SH	Northern caddisfly	Limnephilidae	SH1	5/31/2016	-0.14	-25.21	18	7.01	0.54	0.72	23.47
CR1	ES	SH	Northern caddisfly	Limnephilidae	SH1	5/31/2016	0.09	-25.42	18	4.26	0.62	0.72	13.08
CR1	ES	SH	Shredding stonefly	Plecoptera	SH2	5/31/2016	1.94	-27.4	124	21.48	0.28	0.72	39.52
EL1	SP	GC	Crayfish	Cambaridae	GC2	5/14/2012	2.9	-25.3	1	108.00	4.9	2	213.49
EL1	SP	GC	Crayfish	Cambaridae	GC2	5/14/2012	3.7	-26.4	1	234.00	3.2	2	390.00
EL1	SP	GC	Crayfish	Cambaridae	GC2	5/14/2012	4.5	-26	2	367.00	3.4	2	528.73
EL1	SP	SH	Northern caddisfly	Limnephilidae	SH1	5/14/2012	1.4	-27	9	28.90	0.8	2	87.73
EL1	SP	SH	Northern caddisfly	Limnephilidae	SH1	5/14/2012	1	-26.9	10	21.50	1	2	76.15
EL1	SP	SH	Northern caddisfly	Limnephilidae	SH1	5/14/2012	1.6	-27.1	10	11.40	1	2	32.30
EL1	LS	GC	Crayfish	Cambaridae	GC2	7/9/2012	3.2	-24.8	2	102.00	7.4	2	188.48
EL1	LS	GC	Crayfish	Cambaridae	GC2	7/9/2012	3.5	-25.1	3	306.00	8.8	2	530.82
EL1	LS	GC	Crayfish	Cambaridae	GC2	7/9/2012	3.6	-25	3	304.00	11.9	2	516.80
EL1	LS	GC	Crayfish	Cambaridae	GC2	7/9/2012	4	-25.2	4	250.00	6.4	2	393.52
EL1	LS	GC	Crayfish	Cambaridae	GC2	7/9/2012	4.5	-25.3	4	327.00	2.2	2	471.10
EL1	LS	SH	Northern caddisfly	Limnephilidae	SH1	7/9/2012	3.2	-26.7	13	33.90	1.2	2	62.64
EL1	ES	FC	Brushlegged mayfly	Isonychia	FC1	6/12/2013	3.5	-30.8	11	15.98	0.2	2	27.72

site code	season	feeding group	Common name	Scientific name	feeding group - taxon code	Sample collection date	N15:N14 (per mil)	C13:C12 (per mil)	Individuals in sample	Methylmercury (ng/g dry weight)	Field weight of sample (g)	N15:14 of base consumer for site (per mil)	Calculated methylmercury in trophic position 2.5 consumer (ng/g dry weight)
EL1	ES	GC	Crayfish	Cambaridae	GC2	6/12/2013	5.1	-26.2	5	298.72	1.6	2	390.64
EL1	ES	GC	Crayfish	Cambaridae	GC2	6/12/2013	5.1	-26.1	5	486.84	5	2	636.64
EL1	ES	GC	Crayfish	Cambaridae	GC2	6/12/2013	4	-25.4	2	380.17	6.5	2	598.42
EL1	ES	PR	Darner dragonfly	Aeshnidae	PR1	6/12/2013	6.1	-27.2	1	526.57	0.7	2	596.77
EL1	ES	PR	Darner dragonfly	Aeshnidae	PR1	6/12/2013	6.7	-26.7	3	667.29	0.3	2	700.24
EL1	ES	PR	Darner dragonfly	Aeshnidae	PR1	6/12/2013	9.6	-28.9	3	471.96	0.4	2	364.69
EL1	ES	PR	Dobsonfly	Megaloptera	PR1	6/12/2013	6.7	-25.8	1	551.42	0.4	2	578.65
EL1	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/12/2013	1.1	-27.1	6	27.00	0.7	2	91.81
EL1	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/12/2013	2.8	-28.7	5	26.86	0.6	2	54.36
EL1	ES	SH	Shredding stonefly	Plecoptera	SH2	6/12/2013	5.2	-28.3	68	79.41	0.2	2	102.28
EL1	LS	FC	Net-spinning caddisfly	Hydropsychidae	FC2	8/1/2013	4.6	-27.2	23	233.65	0.3	2	331.00
EL1	LS	FC	Net-spinning caddisfly	Hydropsychidae	FC2	8/1/2013	4.4	-27.4	23	260.34	0.3	2	381.53
EL1	LS	FC	Net-spinning caddisfly	Hydropsychidae	FC2	8/1/2013	4.5	-27.4	23	190.28	0.4	2	274.13
EL1	LS	GC	Crayfish	Cambaridae	GC2	8/1/2013	3.4	-24	1	282.95	4.4	2	501.06
EL1	LS	GC	Crayfish	Cambaridae	GC2	8/1/2013	3.4	-24.5	1	337.68	4.9	2	597.98
EL1	LS	GC	Crayfish	Cambaridae	GC2	8/1/2013	4.3	-24.7	3	77.35	4.7	2	115.35
EL1	LS	GC	Crayfish	Cambaridae	GC2	8/1/2013	3.4	-24	3	221.24	1.5	2	391.78
EL1	LS	PR	Darner dragonfly	Aeshnidae	PR1	8/1/2013	5.3	-27.4	4	350.41	1.3	2	444.55
EL1	LS	PR	Darner dragonfly	Aeshnidae	PR1	8/1/2013	5.6	-27.3	4	483.28	1.4	2	586.84
EL1	LS	PR	Darner dragonfly	Aeshnidae	PR1	8/1/2013	5	-24.8	6	452.64	0.6	2	601.16
EL1	LS	SH	Rolled-winged stoneflies	Leuctridae	SH2	8/1/2013	4.9	-26.3	89	260.09	0.2	2	350.91

site code	season	feeding group	Common name	Scientific name	feeding group - taxon code	Sample collection date	N15:N14 (per mil)	C13:C12 (per mil)	Individuals in sample	Methylmercury (ng/g dry weight)	Field weight of sample (g)	N15:14 of base consumer for site (per mil)	Calculated methylmercury in trophic position 2.5 consumer (ng/g dry weight)
EL1	SP	FC	Brushlegged mayfly	Isonychia	FC1	5/14/2014	4.84	-28.18	23	125.86	0.218	2	171.44
EL1	SP	FC	Net-spinning caddisfly	Hydropsychidae	FC2	5/14/2014	7.5	-26.43	18	488.38	0.306	2	466.43
EL1	SP	GC	Crayfish	Cambaridae	GC2	5/14/2014	5.14	-24.53	1	461.45	1.97	2	599.74
EL1	SP	GC	Crayfish	Cambaridae	GC2	5/14/2014	5.78	-25.07	3	547.12	2.07	2	647.70
EL1	SP	GC	Crayfish	Cambaridae	GC2	5/14/2014	6.12	-24.55	3	597.13	0.694	2	674.95
EL1	SP	PR	Darner dragonfly	Aeshnidae	PR1	5/14/2014	6.77	-27.16	6	1090.06	0.658	2	1134.09
EL1	SP	PR	Darner dragonfly	Aeshnidae	PR1	5/14/2014	6.55	-27.14	2	898.46	1.152	2	960.61
EL1	SP	PR	Darner dragonfly	Aeshnidae	PR1	5/14/2014	6.64	-27.06	2	609.93	1.006	2	644.82
EL1	SP	SH	Northern caddisfly	Limnephilidae	SH1	5/14/2014	2.79	-26.58	5	18.89	0.438	2	38.32
EL1	SP	SH	Northern caddisfly	Limnephilidae	SH1	5/14/2014	2.61	-26.85	5	53.42	0.48	2	113.23
EL1	SP	SH	Northern caddisfly	Limnephilidae	SH1	5/14/2014	2.32	-27.53	6	21.00	0.362	2	47.99
EL1	SP	SH	Rolled-winged stoneflies	Leuctridae	SH2	5/14/2014	4.96	-26.36	40	9.18	0.094	2	12.27
EL1	ES	FC	Net-spinning caddisfly	Hydropsychidae	FC2	6/16/2014	5.62	-26.97	7	198.83	0.09	2	240.75
EL1	ES	GC	Crayfish	Cambaridae	GC2	6/16/2014	4.01	-23.57	1	33.32	10.02	2	52.36
EL1	ES	GC	Crayfish	Cambaridae	GC2	6/16/2014	3.58	-25.36	1	186.88	6.76	2	318.97
EL1	ES	GC	Crayfish	Cambaridae	GC2	6/16/2014	4.76	-24.82	5	262.74	6.95	2	362.54
EL1	ES	PR	Darner dragonfly	Aeshnidae	PR1	6/16/2014	5.94	-27.48	2	391.88	1.42	2	453.81
EL1	ES	PR	Darner dragonfly	Aeshnidae	PR1	6/16/2014	6.07	-26.91	3	450.34	0.48	2	512.43
EL1	ES	PR	Darner dragonfly	Aeshnidae	PR1	6/16/2014	5.59	-27.57	2	394.74	0.42	2	480.01

site code	season	feeding group	Common name	Scientific name	feeding group - taxon code	Sample collection date	N15:N14 (per mil)	C13:C12 (per mil)	Individuals in sample	Methylmercury (ng/g dry weight)	Field weight of sample (g)	N15:14 of base consumer for site (per mil)	Calculated methylmercury in trophic position 2.5 consumer (ng/g dry weight)
EL1	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/16/2014	2.21	-27.54	30	29.15	3.4	2	68.64
EL1	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/16/2014	1.95	-27.74	30	24.00	3.41	2	60.90
EL1	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/16/2014	1.92	-27.62	30	23.96	3.49	2	61.34
EL1	ES	SH	Shredding stonefly	Plecoptera	SH2	6/16/2014	5.08	-26.37	73	56.97	0.13	2	74.74
EL1	LS	GC	Crayfish	Cambaridae	GC2	7/22/2014	3.49	-25.22	1	205.35	6.88	2	356.95
EL1	LS	GC	Crayfish	Cambaridae	GC2	7/22/2014	4.22	-24.87	2	285.09	4.56	2	431.19
EL1	LS	GC	Crayfish	Cambaridae	GC2	7/22/2014	5.2	-25.05	2	305.37	1.18	2	393.28
EL1	LS	PR	Darner dragonfly	Aeshnidae	PR1	7/22/2014	6.05	-27.45	2	307.35	0.75	2	350.67
EL1	LS	PR	Darner dragonfly	Aeshnidae	PR1	7/22/2014	5.76	-27.3	2	390.13	0.65	2	463.14
EL1	LS	PR	Darner dragonfly	Aeshnidae	PR1	7/22/2014	6.31	-26.81	3	332.22	0.18	2	366.27
EL1	ES	FC	Brushlegged mayfly	Isonychia	FC1	6/9/2015	4.78	-27.24	3	---	0.04	2	---
EL1	ES	FC	Net-spinning caddisfly	Hydropsychidae	FC2	6/9/2015	5.59	-26.74	6	---	0.06	2	---
EL1	ES	GC	Crayfish	Cambaridae	GC2	6/9/2015	4.43	-25.42	3	190.56	10.02	2	277.83
EL1	ES	GC	Crayfish	Cambaridae	GC2	6/9/2015	4.75	-25.01	3	397.82	6.78	2	549.83
EL1	ES	GC	Crayfish	Cambaridae	GC2	6/9/2015	4.43	-24.73	3	166.56	6.33	2	242.83
EL1	ES	PR	Darner dragonfly	Aeshnidae	PR1	6/9/2015	7.1	-26.95	1	641.01	0.73	2	641.01
EL1	ES	PR	Darner dragonfly	Aeshnidae	PR1	6/9/2015	6.55	-27.29	1	193.46	0.71	2	206.85
EL1	ES	PR	Dobsonfly	Corydalidae	PR1	6/9/2015	6.59	-26.03	1	---	0.19	2	---
EL1	ES	SC	Flathead mayfly	Heptageniidae	SC1	6/9/2015	4.92	-26.93	20	---	0.28	2	---
EL1	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/9/2015	2.18	-27.06	31	15.28	2.9	2	36.29
EL1	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/9/2015	1.76	-26.95	31	17.24	3.2	2	46.38
EL1	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/9/2015	1.89	-26.95	30	38.22	2.95	2	98.75

site code	season	feeding group	Common name	Scientific name	feeding group - taxon code	Sample collection date	N15:N14 (per mil)	C13:C12 (per mil)	Individuals in sample	Methylmercury (ng/g dry weight)	Field weight of sample (g)	N15:14 of base consumer for site (per mil)	Calculated methylmercury in trophic position 2.5 consumer (ng/g dry weight)
EL1	ES	SH	Shredding stonefly	Plecoptera	SH2	6/9/2015	4.87	-27.01	27	---	0.05	2	---
EL1	LS	FC	Brushlegged mayfly	Isonychia	FC1	7/23/2015	4.32	-28.57	56	---	0.07	2	---
EL1	LS	FC	Net-spinning caddisfly	Hydropsychidae	FC2	7/23/2015	5.5	-26.63	35	---	0.06	2	---
EL1	LS	GC	Caddisfly	Trichoptera	GC1	7/23/2015	3.97	-26.37	19	---	0.16	2	---
EL1	LS	GC	Crayfish	Cambaridae	GC2	7/23/2015	4.27	-24.3	1	226.00	3.35	2	338.80
EL1	LS	GC	Crayfish	Cambaridae	GC2	7/23/2015	5.23	-24.1	1	186.66	2.64	2	239.30
EL1	LS	GC	Crayfish	Cambaridae	GC2	7/23/2015	3.87	-24.24	1	152.13	2.79	2	245.38
EL1	LS	GC	Crayfish	Cambaridae	GC2	7/23/2015	4.02	-24.17	2	260.86	2.32	2	409.09
EL1	LS	GC	Crayfish	Cambaridae	GC2	7/23/2015	4.28	-24.72	1	---	1.14	2	---
EL1	LS	GC	Crayfish	Cambaridae	GC2	7/23/2015	5.1	-24.05	1	296.11	1.39	2	387.22
EL1	LS	PR	Alderfly	Sialidae	PR1	7/23/2015	8.04	-24.76	2	---	0.04	2	---
EL1	LS	PR	Darner dragonfly	Aeshnidae	PR1	7/23/2015	5.56	-27.04	1	344.07	0.4	2	420.20
EL1	LS	PR	Darner dragonfly	Aeshnidae	PR1	7/23/2015	5.11	-26.86	1	304.98	0.22	2	398.20
EL1	LS	PR	Darner dragonfly	Aeshnidae	PR1	7/23/2015	6.87	-27.32	1	586.55	0.31	2	602.87
EL1	LS	PR	Emerald dragonfly	Corduliidae	PR1	7/23/2015	6.6	-26.03	1	209.32	0.38	2	222.40
EL1	LS	SC	Flathead mayfly	Heptageniidae	SC1	7/23/2015	5.39	-26.83	11	---	0.03	2	---
EL1	LS	SH	Northern caddisfly	Limnephilidae	SH1	7/23/2015	1.79	-26.59	6	33.24	0.54	2	88.57
EL1	LS	SH	Shredding stonefly	Plecoptera	SH2	7/23/2015	6.1	-26.15	58	---	0.09	2	---
EL1	ES	FC	Brushlegged mayfly	Isonychia	FC1	6/1/2016	3.27	-27.94	5	---	0.06	2	---

site code	season	feeding group	Common name	Scientific name	feeding group - taxon code	Sample collection date	N15:N14 (per mil)	C13:C12 (per mil)	Individuals in sample	Methylmercury (ng/g dry weight)	Field weight of sample (g)	N15:14 of base consumer for site (per mil)	Calculated methylmercury in trophic position 2.5 consumer (ng/g dry weight)
EL1	ES	FC	Net-spinning caddisfly	Hydropsychidae	FC2	6/1/2016	4.74	-28.01	22	172.28	0.21	2	238.50
EL1	ES	GC	Crayfish	Cambaridae	GC2	6/1/2016	3.81	-24.49	1	324.98	4.83	2	530.20
EL1	ES	GC	Crayfish	Cambaridae	GC2	6/1/2016	4.69	-25.01	1	30.61	1.81	2	42.73
EL1	ES	GC	Crayfish	Cambaridae	GC2	6/1/2016	4.7	-25.35	2	226.10	1.76	2	315.06
EL1	ES	PR	Darner dragonfly	Aeshnidae	PR1	6/1/2016	6.47	-26.68	1	270.19	0.24	2	291.82
EL1	ES	SC	Flathead mayfly	Heptageniidae	SC1	6/1/2016	4.83	-27.73	27	126.06	0.51	2	171.99
EL1	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/1/2016	1.81	-28.11	10	32.60	1.32	2	86.33
EL1	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/1/2016	1.67	-27.41	11	27.08	1.24	2	74.99
EL1	ES	SH	Shredding stonefly	Plecoptera	SH2	6/1/2016	3.64	-27.96	34	---	0.08	2	---
EL1	LS	FC	Net-spinning caddisfly	Hydropsychidae	FC2	7/26/2016	4.72	-27.31	33	86.00	0.37	2	119.44
EL1	LS	GC	Crayfish	Cambaridae	GC2	7/26/2016	4.62	-24.97	1	352.65	4.43	2	497.92
EL1	LS	GC	Crayfish	Cambaridae	GC2	7/26/2016	3.9	-25.35	2	448.52	4.98	2	719.33
EL1	LS	GC	Crayfish	Cambaridae	GC2	7/26/2016	4.13	-24.52	3	281.00	4.24	2	431.92
EL1	LS	PR	Darner dragonfly	Aeshnidae	PR1	7/26/2016	6.12	-27.5	1	653.11	0.41	2	738.22
EL1	LS	PR	Darner dragonfly	Aeshnidae	PR1	7/26/2016	6.36	-27.24	1	788.45	0.38	2	863.64
EL1	LS	PR	Darner dragonfly	Aeshnidae	PR1	7/26/2016	6.11	-26.75	4	972.48	0.64	2	1100.68
EL1	LS	SC	Flathead mayfly	Heptageniidae	SC1	7/26/2016	4.02	-27.54	2	---	0.12	2	---
EL1M	ES	FC	Net-spinning caddisfly	Hydropsychidae	FC2	6/12/2013	4.9	-30.3	5	482.74	0.2	1.27	583.68
EL1M	ES	GC	Crayfish	Cambaridae	GC2	6/12/2013	3.6	-25.9	1	465.19	5	1.27	690.07
EL1M	ES	GC	Crayfish	Cambaridae	GC2	6/12/2013	3.4	-27.2	1	762.96	1	1.27	1172.73
EL1M	ES	PR	Darner dragonfly	Aeshnidae	PR1	6/12/2013	4.2	-30.3	3	895.16	0.3	1.27	1202.03

site code	season	feeding group	Common name	Scientific name	feeding group - taxon code	Sample collection date	N15:N14 (per mil)	C13:C12 (per mil)	Individuals in sample	Methylmercury (ng/g dry weight)	Field weight of sample (g)	N15:14 of base consumer for site (per mil)	Calculated methylmercury in trophic position 2.5 consumer (ng/g dry weight)
EL1M	ES	PR	Spiketail dragonfly	Cordulegastridae	PR1	6/12/2013	4.6	-28.7	5	832.16	0.4	1.27	1051.02
EL1M	ES	PR	Spiketail dragonfly	Cordulegastridae	PR1	6/12/2013	4.4	-28.7	5	771.60	1	1.27	1004.38
EL1M	ES	PR	Spiketail dragonfly	Cordulegastridae	PR1	6/12/2013	3.3	-29.4	1	57.56	0.4	1.27	90.10
EL1M	LS	FC	Net-spinning caddisfly	Hydropsychidae	FC2	8/1/2013	3.5	-30.2	50	1072.83	0.6	1.27	1619.72
EL1M	LS	FC	Net-spinning caddisfly	Hydropsychidae	FC2	8/1/2013	3.7	-30.5	50	1208.96	0.6	1.27	1762.64
EL1M	LS	FC	Net-spinning caddisfly	Hydropsychidae	FC2	8/1/2013	3.7	-30	50	1130.86	0.6	1.27	1648.76
EL1M	LS	GC	Crayfish	Cambaridae	GC2	8/1/2013	3.5	-26.4	1	657.22	1.6	1.27	992.25
EL1M	LS	PR	Darner dragonfly	Aeshnidae	PR1	8/1/2013	3.8	-31.5	3	1073.24	0.8	1.27	1538.36
EL1M	LS	PR	Darner dragonfly	Aeshnidae	PR1	8/1/2013	3.6	-31.2	3	911.61	0.9	1.27	1352.30
EL1M	LS	PR	Darner dragonfly	Aeshnidae	PR1	8/1/2013	3.6	-31.9	3	1181.03	1	1.27	1751.96
EL1M	LS	SH	Rolled-winged stoneflies	Leuctridae	SH2	8/1/2013	1.9	-29.1	95	726.53	0.2	1.27	1532.38
EL1M	SP	FC	Brushlegged mayfly	Isonychia	FC1	5/15/2014	1.31	-33.32	8	242.99	0.178	1.27	600.42
EL1M	SP	FC	Brushlegged mayfly	Isonychia	FC1	5/15/2014	1.26	-33.29	7	253.61	0.16	1.27	635.88
EL1M	SP	FC	Net-spinning caddisfly	Hydropsychidae	FC2	5/15/2014	3.97	-31.23	16	675.25	0.23	1.27	940.92
EL1M	SP	FC	Net-spinning caddisfly	Hydropsychidae	FC2	5/15/2014	4.23	-31.16	16	604.39	0.206	1.27	807.75
EL1M	SP	FC	Net-spinning caddisfly	Hydropsychidae	FC2	5/15/2014	4.4	-31.28	16	657.57	0.23	1.27	855.95
EL1M	SP	GC	Crayfish	Cambaridae	GC2	5/15/2014	3.62	-26.72	1	1010.65	11.66	1.27	1494.00
EL1M	SP	GC	Crayfish	Cambaridae	GC2	5/15/2014	4.07	-28.47	1	658.85	6.95	1.27	903.27
EL1M	SP	GC	Crayfish	Cambaridae	GC2	5/15/2014	4.18	-27.82	1	721.68	2.39	1.27	972.15

site code	season	feeding group	Common name	Scientific name	feeding group - taxon code	Sample collection date	N15:N14 (per mil)	C13:C12 (per mil)	Individuals in sample	Methylmercury (ng/g dry weight)	Field weight of sample (g)	N15:14 of base consumer for site (per mil)	Calculated methylmercury in trophic position 2.5 consumer (ng/g dry weight)
EL1M	SP	PR	Darner dragonfly	Aeshnidae	PR1	5/15/2014	4.38	-31.99	1	1110.34	0.708	1.27	1449.75
EL1M	SP	PR	Darner dragonfly	Aeshnidae	PR1	5/15/2014	4.48	-32.06	2	756.18	0.352	1.27	972.39
EL1M	SP	PR	Spiketail dragonfly	Cordulegastridae	PR1	5/15/2014	4.52	-29.94	4	916.36	0.82	1.27	1171.29
EL1M	SP	SH	Northern caddisfly	Limnephilidae	SH1	5/15/2014	1.57	-29.29	5	283.70	0.496	1.27	651.73
EL1M	SP	SH	Rolled-winged stoneflies	Leuctridae	SH2	5/15/2014	1.26	-32.46	72	211.75	0.108	1.27	530.93
EL1M	ES	FC	Net-spinning caddisfly	Hydropsychidae	FC2	6/16/2014	4.49	-31.11	45	447.63	0.68	1.27	574.75
EL1M	ES	FC	Net-spinning caddisfly	Hydropsychidae	FC2	6/16/2014	4.53	-31.08	45	634.64	0.73	1.27	809.97
EL1M	ES	FC	Net-spinning caddisfly	Hydropsychidae	FC2	6/16/2014	4.63	-31.45	44	534.38	0.67	1.27	671.93
EL1M	ES	GC	Crayfish	Cambaridae	GC2	6/16/2014	2.92	-27.32	1	755.58	10.24	1.27	1271.77
EL1M	ES	GC	Crayfish	Cambaridae	GC2	6/16/2014	5.12	-28.03	1	865.80	9.26	1.27	1015.08
EL1M	ES	GC	Crayfish	Cambaridae	GC2	6/16/2014	4.32	-25.49	1	349.32	4.02	1.27	460.34
EL1M	ES	PR	Darner dragonfly	Aeshnidae	PR1	6/16/2014	4.17	-29.92	1	1201.98	0.72	1.27	1621.71
EL1M	ES	PR	Darner dragonfly	Aeshnidae	PR1	6/16/2014	3.61	-31.99	1	872.74	0.23	1.27	1292.39
EL1M	ES	PR	Dobsonfly	Megaloptera	PR1	6/16/2014	4.7	-28.44	2	801.57	0.54	1.27	997.57
EL1M	ES	PR	Dobsonfly	Megaloptera	PR1	6/16/2014	3.56	-30.34	2	1294.89	0.31	1.27	1934.37
EL1M	ES	PR	Dobsonfly	Megaloptera	PR1	6/16/2014	3.8	-30.17	2	736.95	0.38	1.27	1056.34
EL1M	ES	PR	Spiketail dragonfly	Cordulegastridae	PR1	6/16/2014	4.94	-28.35	3	593.74	0.81	1.27	713.83
EL1M	ES	SH	Shredding stonefly	Plecoptera	SH2	6/16/2014	2.7	-26.84	25	73.18	0.05	1.27	128.79
EL1M	LS	FC	Net-spinning caddisfly	Hydropsychidae	FC2	7/23/2014	3.69	-30.22	50	702.94	0.72	1.27	1026.64

site code	season	feeding group	Common name	Scientific name	feeding group - taxon code	Sample collection date	N15:N14 (per mil)	C13:C12 (per mil)	Individuals in sample	Methylmercury (ng/g dry weight)	Field weight of sample (g)	N15:14 of base consumer for site (per mil)	Calculated methylmercury in trophic position 2.5 consumer (ng/g dry weight)
EL1M	LS	FC	Net-spinning caddisfly	Hydropsychidae	FC2	7/23/2014	3.92	-30.22	50	157.25	0.78	1.27	220.93
EL1M	LS	FC	Net-spinning caddisfly	Hydropsychidae	FC2	7/23/2014	4.25	-30.2	50	678.30	0.74	1.27	903.69
EL1M	LS	GC	Crayfish	Cambaridae	GC2	7/23/2014	3.53	-27.69	1	500.99	0.9	1.27	752.38
EL1M	LS	GC	Crayfish	Cambaridae	GC2	7/23/2014	3.04	-28.87	3	459.81	1.18	1.27	755.97
EL1M	LS	GC	Crayfish	Cambaridae	GC2	7/23/2014	3.93	-27.2	3	458.59	0.94	1.27	643.24
EL1M	LS	PR	Darner dragonfly	Aeshnidae	PR1	7/23/2014	3.55	-30.26	2	705.56	0.61	1.27	1055.86
EL1M	LS	SH	Rolled-winged stoneflies	Leuctridae	SH2	7/23/2014	2.46	-28.64	70	417.84	0.08	1.27	773.77
EL1M	ES	FC	Brushlegged mayfly	Isonychia	FC1	6/9/2015	2.72	-30.18	1	---	0.01	1.27	---
EL1M	ES	FC	Net-spinning caddisfly	Hydropsychidae	FC2	6/9/2015	3.96	-31.68	32	595.53	0.58	1.27	831.20
EL1M	ES	GC	Crayfish	Cambaridae	GC2	6/9/2015	3.3	-27.34	2	455.50	5.13	1.27	713.03
EL1M	ES	GC	Crayfish	Cambaridae	GC2	6/9/2015	2.76	-27.88	3	364.90	4.14	1.27	634.28
EL1M	ES	GC	Crayfish	Cambaridae	GC2	6/9/2015	3.31	-27.49	5	524.81	4.69	1.27	820.01
EL1M	ES	PR	Darner dragonfly	Aeshnidae	PR1	6/9/2015	3.54	-30.68	1	698.33	0.76	1.27	1046.87
EL1M	ES	PR	Dobsonfly	Corydalidae	PR1	6/9/2015	4.39	-29.14	3	792.67	1.09	1.27	1033.39
EL1M	ES	PR	Spiketail dragonfly	Cordulegastridae	PR1	6/9/2015	4.92	-28.71	1	505.38	0.55	1.27	609.33
EL1M	ES	SC	Flathead mayfly	Heptageniidae	SC1	6/9/2015	2.26	-28.8	11	---	0.14	1.27	---
EL1M	ES	SH	Shredding stonefly	Plecoptera	SH2	6/9/2015	5.72	-27	136	173.62	0.34	1.27	187.99
EL1M	LS	FC	Net-spinning caddisfly	Hydropsychidae	FC2	7/22/2015	3.5	-30.43	39	790.85	0.28	1.27	1194.01
EL1M	LS	FC	Net-spinning caddisfly	Hydropsychidae	FC2	7/22/2015	4.18	-30.94	39	919.51	0.32	1.27	1238.64

site code	season	feeding group	Common name	Scientific name	feeding group - taxon code	Sample collection date	N15:N14 (per mil)	C13:C12 (per mil)	Individuals in sample	Methylmercury (ng/g dry weight)	Field weight of sample (g)	N15:14 of base consumer for site (per mil)	Calculated methylmercury in trophic position 2.5 consumer (ng/g dry weight)
EL1M	LS	FC	Net-spinning caddisfly	Hydropsychidae	FC2	7/22/2015	3.59	-30.15	38	508.49	0.24	1.27	755.63
EL1M	LS	SC	Flathead mayfly	Heptageniidae	SC1	7/22/2015	2.36	-28.22	3	---	0.04	1.27	---
EL1M	LS	SH	Rolled-winged stoneflies	Leuctridae	SH2	7/22/2015	4.86	-28.28	78	358.64	0.09	1.27	436.11
EL1M	ES	FC	Black fly	Simuliidae	FC1	6/1/2016	2.49	-30.57	55	---	0.1	1.27	---
EL1M	ES	FC	Net-spinning caddisfly	Hydropsychidae	FC2	6/1/2016	4.17	-30.08	22	498.49	0.48	1.27	672.56
EL1M	ES	FC	Net-spinning caddisfly	Hydropsychidae	FC2	6/1/2016	4.29	-30.06	22	476.30	0.45	1.27	630.62
EL1M	ES	GC	Crayfish	Cambaridae	GC2	6/1/2016	3.63	-28.05	1	868.47	6.01	1.27	1281.60
EL1M	ES	GC	Crayfish	Cambaridae	GC2	6/1/2016	5.05	-27.6	1	759.02	0.93	1.27	898.56
EL1M	ES	PR	Alderfly	Sialidae	PR1	6/1/2016	4.63	-27.9	4	---	0.08	1.27	---
EL1M	ES	PR	Common skimmer	Libellulidae	PR1	6/1/2016	3.17	-30.31	1	404.42	0.24	1.27	648.59
EL1M	ES	PR	Darner dragonfly	Aeshnidae	PR1	6/1/2016	4.96	-30.53	1	1044.09	0.81	1.27	1251.73
EL1M	ES	PR	Darner dragonfly	Aeshnidae	PR1	6/1/2016	4.71	-30.16	1	1039.99	0.86	1.27	1292.39
EL1M	ES	SC	Flathead mayfly	Heptageniidae	SC1	6/1/2016	2.87	-29.21	10	---	0.17	1.27	---
EL1M	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/1/2016	1.32	-29.39	2	---	0.24	1.27	---
EL1M	ES	SH	Shredding stonefly	Plecoptera	SH2	6/1/2016	2.89	-29.16	18	---	0.05	1.27	---
EL1M	LS	FC	Net-spinning caddisfly	Hydropsychidae	FC2	7/27/2016	3.48	-29.6	29	1748.13	0.38	1.27	2648.68
EL1M	LS	FC	Net-spinning caddisfly	Hydropsychidae	FC2	7/27/2016	3.79	-29.55	30	2060.07	0.37	1.27	2957.87
EL1M	LS	FC	Net-spinning caddisfly	Hydropsychidae	FC2	7/27/2016	3.98	-29.61	30	1553.83	0.41	1.27	2161.62
EL1M	LS	GC	Crayfish	Cambaridae	GC2	7/27/2016	3.49	-26.94	2	1276.38	2.44	1.27	1930.47

site code	season	feeding group	Common name	Scientific name	feeding group - taxon code	Sample collection date	N15:N14 (per mil)	C13:C12 (per mil)	Individuals in sample	Methylmercury (ng/g dry weight)	Field weight of sample (g)	N15:14 of base consumer for site (per mil)	Calculated methylmercury in trophic position 2.5 consumer (ng/g dry weight)
EL1M	LS	GC	Crayfish	Cambaridae	GC2	7/27/2016	4.04	-26.04	2	1489.20	2.68	1.27	2051.58
EL1M	LS	GC	Crayfish	Cambaridae	GC2	7/27/2016	3.68	-29.55	1	1202.36	1.8	1.27	1759.04
EL1M	LS	PR	Alderfly	Sialidae	PR1	7/27/2016	3.45	-27.06	8	---	0.16	1.27	---
EL1M	LS	PR	Darner dragonfly	Aeshnidae	PR1	7/27/2016	3.36	-29.44	1	1502.48	0.4	1.27	2326.24
EL1M	LS	PR	Darner dragonfly	Aeshnidae	PR1	7/27/2016	3.43	-29.53	1	2058.71	0.29	1.27	3147.31
EL1M	LS	PR	Darner dragonfly	Aeshnidae	PR1	7/27/2016	3.57	-29.54	1	228.18	0.42	1.27	340.27
EL1M	LS	PR	Spiketail dragonfly	Cordulegastridae	PR1	7/27/2016	3.93	-27.88	1	847.64	0.28	1.27	1188.94
EL1M	LS	SH	Northern caddisfly	Limnephilidae	SH1	7/27/2016	0.91	-29.11	3	249.88	0.33	1.27	698.68
EL1M	LS	SH	Rolled-winged stoneflies	Leuctridae	SH2	7/27/2016	2.88	-28.15	13	---	0.03	1.27	---
EL1P	ES	FC	Brushlegged mayfly	Isonychia	FC1	6/1/2016	3.37	-30.76	8	---	0.13	2.04	---
EL1P	ES	FC	Net-spinning caddisfly	Hydropsychidae	FC2	6/1/2016	5.77	-29.16	10	340.49	0.24	2.04	405.92
EL1P	ES	GC	Crayfish	Cambaridae	GC2	6/1/2016	4.23	-27.21	1	282.39	3.41	2.04	429.40
EL1P	ES	GC	Crayfish	Cambaridae	GC2	6/1/2016	5.19	-28.32	2	117.56	3.7	2.04	152.55
EL1P	ES	GC	Crayfish	Cambaridae	GC2	6/1/2016	1.55	-25.08	2	391.88	1.39	2.04	1144.67
EL1P	ES	PR	Spiketail dragonfly	Cordulegastridae	PR1	6/1/2016	6.85	-26.67	1	279.69	0.95	2.04	289.57
EL1P	ES	PR	Spiketail dragonfly	Cordulegastridae	PR1	6/1/2016	5.9	-27.69	1	372.26	0.57	2.04	435.84
EL1P	ES	PR	Spiketail dragonfly	Cordulegastridae	PR1	6/1/2016	6.09	-26.91	2	580.78	0.27	2.04	662.63
EL1P	ES	SC	Flathead mayfly	Heptageniidae	SC1	6/1/2016	4.11	-31.91	17	291.03	0.72	2.04	452.24
EL1P	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/1/2016	2.03	-29.4	14	41.32	1.62	2.04	103.60

site code	season	feeding group	Common name	Scientific name	feeding group - taxon code	Sample collection date	N15:N14 (per mil)	C13:C12 (per mil)	Individuals in sample	Methylmercury (ng/g dry weight)	Field weight of sample (g)	N15:14 of base consumer for site (per mil)	Calculated methylmercury in trophic position 2.5 consumer (ng/g dry weight)
EL1P	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/1/2016	2.24	-29.48	14	29.85	1.66	2.04	70.47
EL1P	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/1/2016	1.84	-29.17	14	48.24	1.67	2.04	128.13
EL1P	LS	FC	Net-spinning caddisfly	Hydropsychidae	FC2	7/26/2016	4.97	-28.54	36	834.47	0.44	2.04	1120.54
EL1P	LS	FC	Net-spinning caddisfly	Hydropsychidae	FC2	7/26/2016	4.8	-28.7	35	716.40	0.34	2.04	988.54
EL1P	LS	GC	Crayfish	Cambaridae	GC2	7/26/2016	5	-27.04	1	369.11	5.82	2.04	493.31
EL1P	LS	GC	Crayfish	Cambaridae	GC2	7/26/2016	4.17	-26.04	1	572.84	4.28	2.04	880.49
EL1P	LS	GC	Crayfish	Cambaridae	GC2	7/26/2016	4.98	-27.14	3	510.61	5.85	2.04	684.57
EL1P	LS	PR	Darner dragonfly	Aeshnidae	PR1	7/26/2016	5.31	-29.14	1	1081.80	0.43	2.04	1378.60
EL1P	LS	PR	Spiketail dragonfly	Cordulegastridae	PR1	7/26/2016	5.5	-26.67	2	658.38	0.29	2.04	815.78
EL1P	LS	SC	Flathead mayfly	Heptageniidae	SC1	7/26/2016	3.37	-26.91	19	---	0.1	2.04	---
EL1P	LS	SH	Shredding stonefly	Plecoptera	SH2	7/26/2016	4	-27.36	14	---	0.19	2.04	---
EL1R	SP	GC	Crayfish	Cambaridae	GC2	5/15/2012	3.6	-26.9	1	432.00	11.6	2.14	755.56
EL1R	SP	GC	Crayfish	Cambaridae	GC2	5/15/2012	4	-29.3	1	1090.00	6.8	2.14	1761.41
EL1R	SP	GC	Crayfish	Cambaridae	GC2	5/15/2012	3.9	-28.1	1	736.00	7.9	2.14	1212.40
EL1R	SP	PR	Darner dragonfly	Aeshnidae	PR1	5/15/2012	5	-31.4	4	1010.00	1.2	2.14	1371.41
EL1R	SP	PR	Darner dragonfly	Aeshnidae	PR1	5/15/2012	5.4	-31.3	3	930.00	1.2	2.14	1186.94
EL1R	SP	PR	Darner dragonfly	Aeshnidae	PR1	5/15/2012	4.7	-32.5	3	700.00	1.2	2.14	998.32
EL1R	LS	GC	Crayfish	Cambaridae	GC2	7/10/2012	4.8	-28.6	2	1010.00	9	2.14	1416.67
EL1R	LS	GC	Crayfish	Cambaridae	GC2	7/10/2012	4.6	-29	2	828.00	6.6	2.14	1201.02

site code	season	feeding group	Common name	Scientific name	feeding group - taxon code	Sample collection date	N15:N14 (per mil)	C13:C12 (per mil)	Individuals in sample	Methylmercury (ng/g dry weight)	Field weight of sample (g)	N15:14 of base consumer for site (per mil)	Calculated methylmercury in trophic position 2.5 consumer (ng/g dry weight)
EL1R	LS	GC	Crayfish	Cambaridae	GC2	7/10/2012	4.5	-28.6	1	1320.00	8.1	2.14	1947.92
EL1R	LS	GC	Crayfish	Cambaridae	GC2	7/10/2012	5	-28.6	1	1110.00	7.2	2.14	1507.19
EL1R	LS	GC	Crayfish	Cambaridae	GC2	7/10/2012	4.1	-28.7	1	1370.00	8.9	2.14	2172.57
EL1R	LS	PR	Darner dragonfly	Aeshnidae	PR1	7/10/2012	5.1	-31.3	30	1550.00	0.8	2.14	2071.54
EL1R	ES	FC	Net-spinning caddisfly	Hydropsychidae	FC2	6/12/2013	4.5	-30.9	11	508.38	0.3	2.14	750.22
EL1R	ES	FC	Net-spinning caddisfly	Hydropsychidae	FC2	6/12/2013	4.8	-30.7	11	396.12	0.3	2.14	555.62
EL1R	ES	FC	Net-spinning caddisfly	Hydropsychidae	FC2	6/12/2013	4.6	-30.8	12	135.07	0.4	2.14	195.92
EL1R	ES	GC	Crayfish	Cambaridae	GC2	6/12/2013	4.5	-28.9	1	593.65	4.1	2.14	876.04
EL1R	ES	GC	Crayfish	Cambaridae	GC2	6/12/2013	4.3	-30.1	1	941.83	3	2.14	1439.84
EL1R	ES	GC	Crayfish	Cambaridae	GC2	6/12/2013	5.1	-28.5	1	921.19	2.9	2.14	1231.15
EL1R	ES	PR	Darner dragonfly	Aeshnidae	PR1	6/12/2013	4.2	-31.5	6	922.81	0.8	2.14	1436.61
EL1R	ES	PR	Darner dragonfly	Aeshnidae	PR1	6/12/2013	4.6	-31.8	2	511.22	0.8	2.14	741.52
EL1R	ES	PR	Darner dragonfly	Aeshnidae	PR1	6/12/2013	4.3	-31.8	3	468.32	0.9	2.14	715.96
EL1R	LS	FC	Net-spinning caddisfly	Hydropsychidae	FC2	7/30/2013	4.5	-32.4	27	838.18	0.4	2.14	1236.89
EL1R	LS	GC	Crayfish	Cambaridae	GC2	7/30/2013	4.3	-28.3	2	810.83	6.2	2.14	1239.58
EL1R	LS	GC	Crayfish	Cambaridae	GC2	7/30/2013	3.9	-28.9	2	665.45	7.2	2.14	1096.18
EL1R	LS	GC	Crayfish	Cambaridae	GC2	7/30/2013	4.3	-29.4	2	1052.03	7.2	2.14	1608.32
EL1R	LS	PR	Darner dragonfly	Aeshnidae	PR1	7/30/2013	4.7	-33	1	1094.76	0.5	2.14	1561.32
EL1R	LS	PR	Darner dragonfly	Aeshnidae	PR1	7/30/2013	4.6	-32	1	1129.34	0.4	2.14	1638.12

site code	season	feeding group	Common name	Scientific name	feeding group - taxon code	Sample collection date	N15:N14 (per mil)	C13:C12 (per mil)	Individuals in sample	Methylmercury (ng/g dry weight)	Field weight of sample (g)	N15:14 of base consumer for site (per mil)	Calculated methylmercury in trophic position 2.5 consumer (ng/g dry weight)
EL1R	LS	PR	Darner dragonfly	Aeshnidae	PR1	7/30/2013	4.6	-31.3	2	798.52	0.7	2.14	1158.26
EL1R	SP	FC	Brushlegged mayfly	Isonychia	FC1	5/15/2014	1.37	-34.4	11	256.48	0.294	2.14	828.93
EL1R	SP	FC	Net-spinning caddisfly	Hydropsychidae	FC2	5/15/2014	5.36	-32.56	15	718.27	0.448	2.14	922.24
EL1R	SP	FC	Net-spinning caddisfly	Hydropsychidae	FC2	5/15/2014	4.73	-33.08	15	722.36	0.418	2.14	1025.05
EL1R	SP	FC	Net-spinning caddisfly	Hydropsychidae	FC2	5/15/2014	5.42	-32.65	15	1181.90	0.52	2.14	1503.91
EL1R	SP	GC	Crayfish	Cambaridae	GC2	5/15/2014	4.48	-30.29	1	935.10	7.17	2.14	1384.73
EL1R	SP	GC	Crayfish	Cambaridae	GC2	5/15/2014	4.41	-29.63	2	793.97	5.67	2.14	1190.26
EL1R	SP	GC	Crayfish	Cambaridae	GC2	5/15/2014	4.42	-29.9	3	787.29	5.37	2.14	1178.16
EL1R	SP	PR	Darner dragonfly	Aeshnidae	PR1	5/15/2014	4.85	-32.86	1	1222.08	0.692	2.14	1700.11
EL1R	SP	PR	Darner dragonfly	Aeshnidae	PR1	5/15/2014	4.37	-30.57	1	911.63	0.254	2.14	1376.36
EL1R	SP	PR	Darner dragonfly	Aeshnidae	PR1	5/15/2014	4.4	-31.69	1	869.35	0.094	2.14	1305.56
EL1R	SP	PR	Spiketail dragonfly	Cordulegastridae	PR1	5/15/2014	4.73	-30.88	2	2598.85	0.284	2.14	3687.85
EL1R	ES	FC	Net-spinning caddisfly	Hydropsychidae	FC2	6/16/2014	4.77	-32.98	18	215.20	0.58	2.14	303.35
EL1R	ES	FC	Net-spinning caddisfly	Hydropsychidae	FC2	6/16/2014	5.01	-33.1	18	662.25	0.53	2.14	897.78
EL1R	ES	FC	Net-spinning caddisfly	Hydropsychidae	FC2	6/16/2014	5.02	-33.09	17	647.36	0.49	2.14	876.20
EL1R	ES	GC	Crayfish	Cambaridae	GC2	6/16/2014	4.59	-28.61	1	2432.12	9.33	2.14	3533.84
EL1R	ES	GC	Crayfish	Cambaridae	GC2	6/16/2014	4.25	-31.39	2	686.59	4.13	2.14	1059.17
EL1R	ES	GC	Crayfish	Cambaridae	GC2	6/16/2014	4.81	-31.82	1	982.71	3.66	2.14	1376.12
EL1R	ES	PR	Darner dragonfly	Aeshnidae	PR1	6/16/2014	4.3	-33.34	3	501.08	2.37	2.14	766.04

site code	season	feeding group	Common name	Scientific name	feeding group - taxon code	Sample collection date	N15:N14 (per mil)	C13:C12 (per mil)	Individuals in sample	Methylmercury (ng/g dry weight)	Field weight of sample (g)	N15:14 of base consumer for site (per mil)	Calculated methylmercury in trophic position 2.5 consumer (ng/g dry weight)
EL1R	ES	PR	Darner dragonfly	Aeshnidae	PR1	6/16/2014	4.01	-32.53	3	636.17	1.4	2.14	1026.08
EL1R	ES	PR	Darner dragonfly	Aeshnidae	PR1	6/16/2014	4.05	-33.25	1	778.43	0.17	2.14	1246.08
EL1R	ES	PR	Spiketail dragonfly	Cordulegastridae	PR1	6/16/2014	5.37	-30.81	4	777.17	1.1	2.14	996.37
EL1R	LS	FC	Net-spinning caddisfly	Hydropsychidae	FC2	7/23/2014	5.09	-33.17	16	862.58	0.17	2.14	1154.63
EL1R	LS	FC	Net-spinning caddisfly	Hydropsychidae	FC2	7/23/2014	4.79	-32.63	15	1126.28	0.15	2.14	1582.37
EL1R	LS	FC	Net-spinning caddisfly	Hydropsychidae	FC2	7/23/2014	4.66	-33.12	15	1058.22	0.17	2.14	1519.40
EL1R	LS	GC	Crayfish	Cambaridae	GC2	7/23/2014	4.54	-31.67	1	859.32	5.98	2.14	1259.34
EL1R	LS	GC	Crayfish	Cambaridae	GC2	7/23/2014	4.52	-31.76	2	845.70	4.72	2.14	1243.68
EL1R	LS	GC	Crayfish	Cambaridae	GC2	7/23/2014	4.89	-31.1	2	722.78	6.06	2.14	998.96
EL1R	LS	PR	Darner dragonfly	Aeshnidae	PR1	7/23/2014	4.62	-32.06	1	914.99	0.92	2.14	1322.68
EL1R	LS	PR	Darner dragonfly	Aeshnidae	PR1	7/23/2014	4.79	-30.88	5	991.55	0.17	2.14	1393.08
EL1R	LS	PR	Spiketail dragonfly	Cordulegastridae	PR1	7/23/2014	4.8	-30.49	1	672.11	0.6	2.14	942.73
EL1R	ES	FC	Net-spinning caddisfly	Hydropsychidae	FC2	6/9/2015	4.59	-32.29	22	670.43	0.66	2.14	974.13
EL1R	ES	FC	Net-spinning caddisfly	Hydropsychidae	FC2	6/9/2015	4.97	-32.23	22	661.10	0.63	2.14	901.98
EL1R	ES	FC	Net-spinning caddisfly	Hydropsychidae	FC2	6/9/2015	4.69	-32.1	22	818.45	0.7	2.14	1169.21
EL1R	ES	GC	Crayfish	Cambaridae	GC2	6/9/2015	3.83	-29.37	3	---	9.77	2.14	---
EL1R	ES	GC	Crayfish	Cambaridae	GC2	6/9/2015	4.11	-30.41	3	---	16.65	2.14	---
EL1R	ES	GC	Crayfish	Cambaridae	GC2	6/9/2015	3.8	-30.01	3	---	8.88	2.14	---
EL1R	ES	PR	Darner dragonfly	Aeshnidae	PR1	6/9/2015	4.31	-32.49	2	675.35	1.29	2.14	1030.61

site code	season	feeding group	Common name	Scientific name	feeding group - taxon code	Sample collection date	N15:N14 (per mil)	C13:C12 (per mil)	Individuals in sample	Methylmercury (ng/g dry weight)	Field weight of sample (g)	N15:14 of base consumer for site (per mil)	Calculated methylmercury in trophic position 2.5 consumer (ng/g dry weight)
EL1R	ES	PR	Darner dragonfly	Aeshnidae	PR1	6/9/2015	3.59	-32.24	2	478.52	1.15	2.14	838.64
EL1R	ES	PR	Darner dragonfly	Aeshnidae	PR1	6/9/2015	3.79	-32.7	2	546.14	1.57	2.14	919.24
EL1R	ES	PR	Dobsonfly	Corydalidae	PR1	6/9/2015	4.79	-32.31	2	---	0.72	2.14	---
EL1R	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/9/2015	3.13	-31.76	5	25.54	0.56	2.14	49.46
EL1R	ES	SH	Shredding stonefly	Plecoptera	SH2	6/9/2015	2.67	-31.45	28	---	0.07	2.14	---
EL1R	LS	FC	Net-spinning caddisfly	Hydropsychidae	FC2	7/22/2015	3.37	-29.72	2	---	0.03	2.14	---
EL1R	LS	PR	Alderfly	Sialidae	PR1	7/22/2015	2.94	-28.16	5	307.88	0.24	2.14	623.10
EL1R	LS	PR	Darner dragonfly	Aeshnidae	PR1	7/22/2015	5.86	-33.33	1	60.26	1.06	2.14	71.94
EL1R	LS	PR	Darner dragonfly	Aeshnidae	PR1	7/22/2015	5.04	-30.38	2	426.58	0.52	2.14	575.54
EL1R	LS	PR	Darner dragonfly	Aeshnidae	PR1	7/22/2015	4.87	-30.81	3	226.30	0.58	2.14	313.79
EL1R	LS	PR	Darner dragonfly	Aeshnidae	PR1	7/22/2015	5.13	-31.52	3	248.42	0.49	2.14	330.44
EL1R	LS	PR	Dobsonfly	Corydalidae	PR1	7/22/2015	5.72	-31.65	1	1669.48	0.57	2.14	2033.03
EL1R	LS	PR	Emerald dragonfly	Corduliidae	PR1	7/22/2015	4.46	-30.17	1	823.71	0.25	2.14	1224.05
EL1R	LS	PR	Emerald dragonfly	Corduliidae	PR1	7/22/2015	4.8	-31.72	1	618.51	0.35	2.14	867.54
EL1R	LS	PR	Emerald dragonfly	Corduliidae	PR1	7/22/2015	4.71	-30.28	1	1076.15	0.37	2.14	1532.21
EL1R	ES	FC	Black fly	Simuliidae	FC1	6/1/2016	2.9	-32.59	80	---	0.2	2.14	---
EL1R	ES	GC	Crayfish	Cambaridae	GC2	6/1/2016	4.58	-29.62	1	709.40	1.97	2.14	1032.52
EL1R	ES	PR	Alderfly	Sialidae	PR1	6/1/2016	4.7	-28.39	6	---	0.15	2.14	---
EL1R	ES	PR	Common skimmer	Libellulidae	PR1	6/1/2016	3.54	-30.13	1	---	0.2	2.14	---

site code	season	feeding group	Common name	Scientific name	feeding group - taxon code	Sample collection date	N15:N14 (per mil)	C13:C12 (per mil)	Individuals in sample	Methylmercury (ng/g dry weight)	Field weight of sample (g)	N15:14 of base consumer for site (per mil)	Calculated methylmercury in trophic position 2.5 consumer (ng/g dry weight)
EL1R	ES	PR	Darner dragonfly	Aeshnidae	PR1	6/1/2016	4.1	-31.94	1	447.27	0.5	2.14	709.30
EL1R	ES	PR	Large caddisflies	Phryganeidae	PR1	6/1/2016	4.11	-31.68	7	364.81	0.58	2.14	577.45
EL1R	LS	FC	Net-spinning caddisfly	Hydropsychidae	FC2	7/27/2016	4.16	-31.33	33	1354.53	0.53	2.14	2124.26
EL1R	LS	FC	Net-spinning caddisfly	Hydropsychidae	FC2	7/27/2016	4.23	-31.29	32	1845.71	0.5	2.14	2857.66
EL1R	LS	FC	Net-spinning caddisfly	Hydropsychidae	FC2	7/27/2016	4.27	-31.38	33	731.52	0.54	2.14	1124.39
EL1R	LS	GC	Crayfish	Cambaridae	GC2	7/27/2016	3.54	-28.58	2	361.62	7.44	2.14	640.37
EL1R	LS	GC	Crayfish	Cambaridae	GC2	7/27/2016	3.79	-28.44	2	471.23	7.49	2.14	793.15
EL1R	LS	GC	Crayfish	Cambaridae	GC2	7/27/2016	4.26	-28.59	2	1269.24	3.48	2.14	1954.44
EL1R	LS	PR	Alderfly	Sialidae	PR1	7/27/2016	4.04	-28.54	2	---	0.12	2.14	---
EL1R	LS	PR	Darner dragonfly	Aeshnidae	PR1	7/27/2016	4.41	-28.69	7	1537.56	0.57	2.14	2304.99
EL1R	LS	PR	Darner dragonfly	Aeshnidae	PR1	7/27/2016	4.36	-28.86	6	1445.03	0.42	2.14	2185.54
EL1R	LS	PR	Large caddisflies	Phryganeidae	PR1	7/27/2016	4.02	-29.27	2	---	0.09	2.14	---
EL1R	LS	PR	Spiketail dragonfly	Cordulegastridae	PR1	7/27/2016	3.83	-28.65	2	507.12	0.47	2.14	846.87
EL1R	LS	SH	Shredding stonefly	Plecoptera	SH2	7/27/2016	3.38	-29.49	32	---	0.05	2.14	---
EL2R	SP	GC	Crayfish	Cambaridae	GC2	5/15/2012	3	-24.6	1	63.40	3.7	1.32	106.08
EL2R	SP	SH	Northern caddisfly	Limnephilidae	SH1	5/15/2012	1.3	-30.3	17	10.00	1.3	1.32	25.15
EL2R	SP	SH	Northern caddisfly	Limnephilidae	SH1	5/15/2012	1.8	-29.6	16	7.87	1.5	1.32	17.24
EL2R	SP	SH	Northern caddisfly	Limnephilidae	SH1	5/15/2012	1.8	-28.8	17	11.30	1.2	1.32	24.76
EL2R	LS	GC	Crayfish	Cambaridae	GC2	7/10/2012	3	-25.2	3	86.80	9.2	1.32	145.24

site code	season	feeding group	Common name	Scientific name	feeding group - taxon code	Sample collection date	N15:N14 (per mil)	C13:C12 (per mil)	Individuals in sample	Methylmercury (ng/g dry weight)	Field weight of sample (g)	N15:14 of base consumer for site (per mil)	Calculated methylmercury in trophic position 2.5 consumer (ng/g dry weight)
EL2R	LS	GC	Crayfish	Cambaridae	GC2	7/10/2012	2.7	-25.8	3	57.70	8.1	1.32	102.60
EL2R	LS	GC	Crayfish	Cambaridae	GC2	7/10/2012	3.2	-25.3	3	52.60	9.4	1.32	84.68
EL2R	LS	SH	Northern caddisfly	Limnephilidae	SH1	7/10/2012	2	-27.9	31	28.40	0.9	1.32	59.17
EL2R	ES	FC	Brushlegged mayfly	Isonychia	FC1	6/13/2013	2.8	-30.1	13	16.21	0.2	1.32	28.23
EL2R	ES	GC	Crayfish	Cambaridae	GC2	6/13/2013	2.4	-25.4	1	61.90	5.3	1.32	117.44
EL2R	ES	GC	Crayfish	Cambaridae	GC2	6/13/2013	3.1	-25.9	1	85.73	2.7	1.32	140.67
EL2R	ES	GC	Crayfish	Cambaridae	GC2	6/13/2013	2.7	-24.9	1	75.80	2.1	1.32	134.78
EL2R	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/13/2013	1	-28.4	5	8.78	0.6	1.32	24.23
EL2R	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/13/2013	0.4	-27.6	5	14.13	0.6	1.32	48.42
EL2R	ES	SH	Shredding stonefly	Plecoptera	SH2	6/13/2013	3.1	-28.4	154	7.19	0.4	1.32	11.80
EL2R	LS	GC	Crayfish	Cambaridae	GC2	7/31/2013	2.7	-24.1	1	47.58	4.2	1.32	84.61
EL2R	LS	GC	Crayfish	Cambaridae	GC2	7/31/2013	4.3	-25.3	3	72.36	1	1.32	96.40
EL2R	LS	GC	Crayfish	Cambaridae	GC2	7/31/2013	4.4	-25.1	2	85.31	1	1.32	111.90
EL2R	LS	SH	Shredding stonefly	Plecoptera	SH2	7/31/2013	3.1	-28.4	108	32.52	0.2	1.32	53.36
EL2R	SP	GC	Eurylophella mayfly	Eurylophella	GC1	5/15/2014	2.9	-29.41	13	24.18	0.126	1.32	41.27
EL2R	SP	GC	Eurylophella mayfly	Eurylophella	GC1	5/15/2014	2.98	-29.41	13	20.83	0.128	1.32	34.99
EL2R	SP	GC	Eurylophella mayfly	Eurylophella	GC1	5/15/2014	2.83	-29.15	13	25.99	0.116	1.32	44.99
EL2R	SP	GC	Crayfish	Cambaridae	GC2	5/15/2014	4.11	-27.54	1	87.94	1.802	1.32	120.76
EL2R	SP	GC	Crayfish	Cambaridae	GC2	5/15/2014	3.68	-25.35	1	53.05	1.55	1.32	78.28

site code	season	feeding group	Common name	Scientific name	feeding group - taxon code	Sample collection date	N15:N14 (per mil)	C13:C12 (per mil)	Individuals in sample	Methylmercury (ng/g dry weight)	Field weight of sample (g)	N15:14 of base consumer for site (per mil)	Calculated methylmercury in trophic position 2.5 consumer (ng/g dry weight)
EL2R	SP	GC	Crayfish	Cambaridae	GC2	5/15/2014	3.38	-26.5	1	85.54	13.42	1.32	133.16
EL2R	SP	PR	Perlodid stoneflies	Perlodidae	PR1	5/15/2014	6.41	-28.94	9	60.58	0.19	1.32	60.65
EL2R	SP	SH	Northern caddisfly	Limnephilidae	SH1	5/15/2014	1.08	-29.34	7	11.23	0.35	1.32	30.22
EL2R	SP	SH	Northern caddisfly	Limnephilidae	SH1	5/15/2014	1.07	-29.07	7	13.29	0.396	1.32	35.86
EL2R	SP	SH	Northern caddisfly	Limnephilidae	SH1	5/15/2014	1.05	-28.35	7	9.92	0.452	1.32	26.94
EL2R	SP	SH	Shredding stonefly	Plecoptera	SH2	5/15/2014	4.27	-27.09	77	26.52	0.098	1.32	35.50
EL2R	ES	FC	Brushlegged mayfly	Isonychia	FC1	6/17/2014	2.61	-30.21	10	28.83	0.13	1.32	52.26
EL2R	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/17/2014	1.33	-28.64	10	10.47	0.94	1.32	26.10
EL2R	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/17/2014	0.93	-28.39	10	11.71	1.07	1.32	33.07
EL2R	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/17/2014	1.06	-28.06	9	12.35	0.96	1.32	33.44
EL2R	ES	SH	Shredding stonefly	Plecoptera	SH2	6/17/2014	3.09	-30.09	124	21.45	0.3	1.32	35.26
EL2R	LS	GC	Crayfish	Cambaridae	GC2	7/23/2014	3.78	-26.26	1	91.78	5.44	1.32	133.13
EL2R	LS	GC	Crayfish	Cambaridae	GC2	7/23/2014	4.27	-25.23	2	115.90	2.03	1.32	155.14
EL2R	LS	PR	Spiketail dragonfly	Cordulegastridae	PR1	7/23/2014	4.74	-28.62	1	76.55	0.55	1.32	95.40
EL2R	LS	SH	Shredding stonefly	Plecoptera	SH2	7/23/2014	3.82	-27.11	26	30.20	0.06	1.32	43.51
EL2R	ES	FC	Brushlegged mayfly	Isonychia	FC1	6/10/2015	4.36	-26.43	17	14.98	0.3	1.32	19.78
EL2R	ES	GC	Crane fly	Limoniinae	GC1	6/10/2015	2.68	-24.8	1	---	0.02	1.32	---
EL2R	ES	GC	Crayfish	Cambaridae	GC2	6/10/2015	4.52	-25.29	1	69.36	2.44	1.32	89.32
EL2R	ES	GC	Crayfish	Cambaridae	GC2	6/10/2015	4.53	-27	1	75.66	0.93	1.32	97.29

site code	season	feeding group	Common name	Scientific name	feeding group - taxon code	Sample collection date	N15:N14 (per mil)	C13:C12 (per mil)	Individuals in sample	Methylmercury (ng/g dry weight)	Field weight of sample (g)	N15:14 of base consumer for site (per mil)	Calculated methylmercury in trophic position 2.5 consumer (ng/g dry weight)
EL2R	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/10/2015	0.96	-27.7	5	7.57	0.45	1.32	21.16
EL2R	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/10/2015	1.16	-28.05	5	3.30	0.38	1.32	8.64
EL2R	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/10/2015	2.72	-29.13	5	10.95	0.39	1.32	19.38
EL2R	ES	SH	Shredding stonefly	Plecoptera	SH2	6/10/2015	3.91	-26.95	121	16.84	0.3	1.32	23.89
EL2R	LS	FC	Brushlegged mayfly	Isonychia	FC1	7/21/2015	2.5	-33.34	16	---	0.03	1.32	---
EL2R	LS	FC	Net-spinning caddisfly	Hydropsychidae	FC2	7/21/2015	2.53	-27.55	2	---	0.03	1.32	---
EL2R	LS	GC	Crayfish	Cambaridae	GC2	7/21/2015	3.62	-27.25	2	4.32	5.7	1.32	6.44
EL2R	LS	GC	Crayfish	Cambaridae	GC2	7/21/2015	3.86	-27.59	2	93.86	3.34	1.32	134.32
EL2R	LS	GC	Crayfish	Cambaridae	GC2	7/21/2015	4.68	-26.27	3	123.12	4.41	1.32	154.82
EL2R	LS	SH	Caddisfly	Trichoptera	SH1	7/21/2015	1.5	-28.62	11	0.79	1.09	1.32	1.87
EL2R	LS	SH	Shredding stonefly	Plecoptera	SH2	7/21/2015	3.16	-29.53	24	---	0.05	1.32	---
ER1	ES	FC	Brushlegged mayfly	Isonychia	FC1	6/12/2013	2.6	-32.9	24	283.61	0.4	2.8	753.33
ER1	ES	PR	Darner dragonfly	Aeshnidae	PR1	6/12/2013	5.8	-30.2	5	565.70	1.6	2.8	751.32
ER1	ES	PR	Darner dragonfly	Aeshnidae	PR1	6/12/2013	5.7	-29.9	5	636.72	1.9	2.8	859.07
ER1	ES	PR	Darner dragonfly	Aeshnidae	PR1	6/12/2013	5.6	-29.8	5	473.65	1.4	2.8	649.36
ER1	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/12/2013	3.4	-31.1	6	181.65	0.7	2.8	386.01
ER1	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/12/2013	3.7	-30.9	6	190.20	0.8	2.8	375.98
ER1	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/12/2013	3.7	-30.9	6	197.32	0.8	2.8	390.04
ER1	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/12/2013	3.1	-31	6	206.17	0.6	2.8	473.64

site code	season	feeding group	Common name	Scientific name	feeding group - taxon code	Sample collection date	N15:N14 (per mil)	C13:C12 (per mil)	Individuals in sample	Methylmercury (ng/g dry weight)	Field weight of sample (g)	N15:14 of base consumer for site (per mil)	Calculated methylmercury in trophic position 2.5 consumer (ng/g dry weight)
ER1	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/12/2013	4.7	-29.4	5	233.40	0.2	2.8	374.33
ER1	LS	GC	Crayfish	Cambaridae	GC2	7/30/2013	5.1	-28.2	1	639.34	3.8	2.8	953.40
ER1	LS	GC	Crayfish	Cambaridae	GC2	7/30/2013	5.1	-29.4	2	716.14	1.2	2.8	1067.93
ER1	LS	GC	Crayfish	Cambaridae	GC2	7/30/2013	5.6	-29	2	701.54	1.2	2.8	961.79
ER1	LS	PR	Darner dragonfly	Aeshnidae	PR1	7/30/2013	5.3	-29.9	3	599.26	1	2.8	863.34
ER1	LS	PR	Darner dragonfly	Aeshnidae	PR1	7/30/2013	5.8	-30.3	3	912.86	0.8	2.8	1212.39
ER1	LS	PR	Darner dragonfly	Aeshnidae	PR1	7/30/2013	5.4	-29.2	3	1192.70	0.9	2.8	1689.66
ER1	SP	FC	Brushlegged mayfly	Isonychia	FC1	5/14/2014	2.78	-32.21	30	151.73	0.578	2.8	381.56
ER1	SP	GC	Crayfish	Cambaridae	GC2	5/14/2014	2.18	-24.28	1	140.46	12.852	2.8	429.47
ER1	SP	GC	Crayfish	Cambaridae	GC2	5/14/2014	3.13	-25.18	1	178.21	1.786	2.8	406.10
ER1	SP	PR	Alderfly	Sialidae	PR1	5/14/2014	5.61	-28.68	31	57.19	0.25	2.8	78.28
ER1	SP	PR	Darner dragonfly	Aeshnidae	PR1	5/14/2014	5.7	-29.9	1	10.22	0.422	2.8	13.79
ER1	SP	PR	Darner dragonfly	Aeshnidae	PR1	5/14/2014	5.93	-31.24	3	691.21	0.328	2.8	899.74
ER1	SP	PR	Large caddisflies	Phryganeidae	PR1	5/14/2014	5.94	-30.3	8	295.97	0.772	2.8	384.67
ER1	SP	SH	Northern caddisfly	Limnephilidae	SH1	5/14/2014	4.14	-30.64	4	162.67	0.082	2.8	291.71
ER1	ES	FC	Mayflies	Ephemeroptera	FC1	6/17/2014	2.95	-32.53	31	226.20	0.18	2.8	541.60
ER1	ES	GC	Crayfish	Cambaridae	GC2	6/17/2014	3.05	-25.52	1	125.95	5.07	2.8	293.31
ER1	ES	GC	Crayfish	Cambaridae	GC2	6/17/2014	2.49	-24.84	1	98.94	2.76	2.8	272.17
ER1	ES	PR	Darner dragonfly	Aeshnidae	PR1	6/17/2014	5.46	-30.97	3	435.26	1.74	2.8	610.52

site code	season	feeding group	Common name	Scientific name	feeding group - taxon code	Sample collection date	N15:N14 (per mil)	C13:C12 (per mil)	Individuals in sample	Methylmercury (ng/g dry weight)	Field weight of sample (g)	N15:14 of base consumer for site (per mil)	Calculated methylmercury in trophic position 2.5 consumer (ng/g dry weight)
ER1	ES	PR	Darner dragonfly	Aeshnidae	PR1	6/17/2014	5.6	-30.24	4	1036.13	1.38	2.8	1420.50
ER1	ES	PR	Darner dragonfly	Aeshnidae	PR1	6/17/2014	5.31	-30.83	3	390.75	0.55	2.8	561.99
ER1	ES	PR	Spiketail dragonfly	Cordulegastridae	PR1	6/17/2014	5.76	-29.78	5	679.92	1.03	2.8	908.70
ER1	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/17/2014	3.38	-31.33	4	51.09	0.46	2.8	109.11
ER1	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/17/2014	3.52	-31.87	4	182.29	0.48	2.8	376.08
ER1	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/17/2014	3.39	-30.96	3	132.04	0.4	2.8	281.28
ER1	ES	SH	Rolled-winged stoneflies	Leuctridae	SH2	6/17/2014	4.41	-27.21	64	86.29	0.12	2.8	146.39
ER1	LS	GC	Crayfish	Cambaridae	GC2	7/23/2014	2.85	-24.57	1	149.00	4.78	2.8	367.10
ER1	LS	GC	Crayfish	Cambaridae	GC2	7/23/2014	3.87	-25.38	3	139.87	3.71	2.8	265.97
ER1	LS	PR	Darner dragonfly	Aeshnidae	PR1	7/23/2014	5.58	-31.48	14	417.27	0.2	2.8	573.92
ER1	LS	PR	Darner dragonfly	Aeshnidae	PR1	7/23/2014	5.45	-29.71	2	488.61	0.67	2.8	686.47
ER1	LS	PR	Spiketail dragonfly	Cordulegastridae	PR1	7/23/2014	6.45	-28.63	2	1224.45	0.2	2.8	1476.29
ER1	ES	FC	Brushlegged mayfly	Isonychia	FC1	6/10/2015	3.43	-32.27	13	---	0.12	2.8	---
ER1	ES	FC	Brushlegged mayfly	Isonychia	FC1	6/10/2015	4.71	-30.43	1	234.80	1.38	2.8	375.85
ER1	ES	GC	Crayfish	Cambaridae	GC2	6/10/2015	3.3	-25.11	1	164.40	3.36	2.8	358.30
ER1	ES	GC	Crayfish	Cambaridae	GC2	6/10/2015	5.75	-26.92	1	388.34	2.81	2.8	519.83
ER1	ES	GC	Crayfish	Cambaridae	GC2	6/10/2015	5.93	-28.59	1	626.21	2.11	2.8	815.12
ER1	ES	PR	Darner dragonfly	Aeshnidae	PR1	6/10/2015	5.28	-30.59	4	30.88	1.14	2.8	44.64
ER1	ES	PR	Darner dragonfly	Aeshnidae	PR1	6/10/2015	5.72	-30.24	4	517.36	1.05	2.8	695.81

site code	season	feeding group	Common name	Scientific name	feeding group - taxon code	Sample collection date	N15:N14 (per mil)	C13:C12 (per mil)	Individuals in sample	Methylmercury (ng/g dry weight)	Field weight of sample (g)	N15:14 of base consumer for site (per mil)	Calculated methylmercury in trophic position 2.5 consumer (ng/g dry weight)
ER1	ES	PR	Darner dragonfly	Aeshnidae	PR1	6/10/2015	5.66	-31.82	4	508.92	1.31	2.8	691.02
ER1	ES	PR	Darner dragonfly	Aeshnidae	PR1	6/10/2015	5.66	-30.23	6	826.89	0.66	2.8	1122.77
ER1	ES	PR	Emerald dragonfly	Corduliidae	PR1	6/10/2015	6.07	-28.85	1	699.16	0.35	2.8	890.98
ER1	ES	SC	Flathead mayfly	Heptageniidae	SC1	6/10/2015	2.98	-32.38	104	286.86	0.53	2.8	681.08
ER1	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/10/2015	4.17	-30.4	11	230.97	0.75	2.8	411.58
ER1	LS	FC	Net-spinning caddisfly	Hydropsychidae	FC2	7/22/2015	2.8	-25.2	47	---	0.15	2.8	---
ER1	LS	GC	Crayfish	Cambaridae	GC2	7/22/2015	4.48	-28.64	1	66.37	3.43	2.8	111.05
ER1	LS	GC	Crayfish	Cambaridae	GC2	7/22/2015	2.57	-24.15	1	241.36	3.17	2.8	647.19
ER1	LS	GC	Crayfish	Cambaridae	GC2	7/22/2015	5.71	-30.03	1	30.46	2.51	2.8	41.03
ER1	LS	GC	Crayfish	Cambaridae	GC2	7/22/2015	4.93	-30.91	1	171.25	3.92	2.8	263.22
ER1	LS	PR	Alderfly	Sialidae	PR1	7/22/2015	2.08	-24.71	8	177.42	0.16	2.8	562.72
ER1	LS	PR	Common skimmer	Libellulidae	PR1	7/22/2015	4.61	-31.4	2	511.53	0.52	2.8	834.55
ER1	LS	PR	Darner dragonfly	Aeshnidae	PR1	7/22/2015	5.53	-30.03	1	557.37	0.41	2.8	772.86
ER1	LS	PR	Darner dragonfly	Aeshnidae	PR1	7/22/2015	5.45	-29.86	16	126.56	0.14	2.8	177.81
ER1	LS	PR	Emerald dragonfly	Corduliidae	PR1	7/22/2015	5.19	-29.04	1	687.54	0.48	2.8	1009.34
ER1	LS	PR	Emerald dragonfly	Corduliidae	PR1	7/22/2015	5.2	-28.02	2	460.42	0.19	2.8	674.76
ER1	ES	FC	Brushlegged mayfly	Isonychia	FC1	6/1/2016	2.67	-32.35	7	---	0.15	2.8	---
ER1	ES	GC	Crayfish	Cambaridae	GC2	6/1/2016	5.06	-28.42	1	856.39	2.48	2.8	1286.10
ER1	ES	GC	Crayfish	Cambaridae	GC2	6/1/2016	5.28	-28.6	2	66.40	3.24	2.8	95.99
ER1	ES	GC	Crayfish	Cambaridae	GC2	6/1/2016	5.18	-28.91	2	179.17	2.96	2.8	263.49

site code	season	feeding group	Common name	Scientific name	feeding group - taxon code	Sample collection date	N15:N14 (per mil)	C13:C12 (per mil)	Individuals in sample	Methylmercury (ng/g dry weight)	Field weight of sample (g)	N15:14 of base consumer for site (per mil)	Calculated methylmercury in trophic position 2.5 consumer (ng/g dry weight)
ER1	ES	PR	Alderfly	Sialidae	PR1	6/1/2016	5.28	-28.96	31	129.92	0.36	2.8	187.81
ER1	ES	PR	Common skimmer	Libellulidae	PR1	6/1/2016	4.94	-30.83	4	912.21	0.44	2.8	1399.60
ER1	ES	PR	Common skimmer	Libellulidae	PR1	6/1/2016	5.04	-31.91	4	826.42	1.8	2.8	1245.50
ER1	ES	PR	Darner dragonfly	Aeshnidae	PR1	6/1/2016	5.45	-30.83	2	717.32	0.31	2.8	1007.80
ER1	ES	PR	Darner dragonfly	Aeshnidae	PR1	6/1/2016	5.33	-31	2	650.14	0.41	2.8	931.90
ER1	ES	PR	Predaceous diving beetle	Dytiscidae	PR1	6/1/2016	4.86	-30.45	1	---	0.32	2.8	---
ER1	ES	PR	Spiketail dragonfly	Cordulegastridae	PR1	6/1/2016	5.56	-30.03	4	532.61	0.52	2.8	734.93
ER1	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/1/2016	2.48	-32.91	4	150.82	0.49	2.8	416.22
ER1	LS	GC	Crayfish	Cambaridae	GC2	7/27/2016	5.57	-27.79	1	218.86	3.45	2.8	301.51
ER1	LS	GC	Crayfish	Cambaridae	GC2	7/27/2016	5.67	-27.51	1	725.52	3.46	2.8	983.57
ER1	LS	GC	Crayfish	Cambaridae	GC2	7/27/2016	5.03	-28.5	1	77.52	2.25	2.8	117.04
ER1	LS	PR	Alderfly	Sialidae	PR1	7/27/2016	5.2	-27.77	12	88.61	0.31	2.8	129.86
ER1	LS	PR	Alderfly	Sialidae	PR1	7/27/2016	5.33	-28.27	12	115.86	0.24	2.8	166.07
ER1	LS	PR	Alderfly	Sialidae	PR1	7/27/2016	5.1	-28.68	11	64.98	0.23	2.8	96.89
ER1	LS	PR	Common skimmer	Libellulidae	PR1	7/27/2016	5.67	-29.22	5	103.11	0.78	2.8	139.78
ER1	LS	PR	Crane fly	Tipulidae	PR1	7/27/2016	6.75	-28.08	1	---	0.05	2.8	---
ER1	LS	PR	Darner dragonfly	Aeshnidae	PR1	7/27/2016	5.46	-31.16	1	721.58	0.91	2.8	1012.12
ER1	LS	PR	Darner dragonfly	Aeshnidae	PR1	7/27/2016	5.94	-30.39	2	1122.32	0.36	2.8	1458.67
ER1	LS	PR	Spiketail dragonfly	Cordulegastridae	PR1	7/27/2016	5.46	-30.68	1	855.64	0.81	2.8	1200.15
ER1	LS	PR	Spiketail dragonfly	Cordulegastridae	PR1	7/27/2016	5.45	-29.09	3	586.95	0.49	2.8	824.64
ER2	SP	GC	Crayfish	Cambaridae	GC2	5/15/2012	4.1	-25.2	1	35.20	4.7	2.36	58.21

site code	season	feeding group	Common name	Scientific name	feeding group - taxon code	Sample collection date	N15:N14 (per mil)	C13:C12 (per mil)	Individuals in sample	Methylmercury (ng/g dry weight)	Field weight of sample (g)	N15:14 of base consumer for site (per mil)	Calculated methylmercury in trophic position 2.5 consumer (ng/g dry weight)
ER2	SP	SH	Northern caddisfly	Limnephilidae	SH1	5/15/2012	3.3	-30.1	10	18.00	0.9	2.36	35.25
ER2	SP	SH	Northern caddisfly	Limnephilidae	SH1	5/15/2012	2.7	-28.4	10	6.98	0.8	2.36	15.86
ER2	SP	SH	Northern caddisfly	Limnephilidae	SH1	5/15/2012	2.8	-29.6	10	3.63	0.8	2.36	8.04
ER2	LS	GC	Aquatic sowbug	Asellus aquaticus	GC1	7/9/2012	5.1	-26.3	39	137.00	0.6	2.36	189.66
ER2	LS	GC	Crayfish	Cambaridae	GC2	7/9/2012	3	-24.5	1	75.20	4.7	2.36	158.22
ER2	LS	GC	Crayfish	Cambaridae	GC2	7/9/2012	4.8	-24.9	2	125.00	3.5	2.36	181.93
ER2	LS	SH	Northern caddisfly	Limnephilidae	SH1	7/9/2012	3.2	-27.7	44	24.80	1.2	2.36	49.72
ER2	LS	SH	Northern caddisfly	Limnephilidae	SH1	7/9/2012	3.2	-27.6	47	27.60	1.3	2.36	55.33
ER2	LS	SH	Northern caddisfly	Limnephilidae	SH1	7/9/2012	3.6	-27.8	43	26.80	1.2	2.36	49.09
ER2	ES	GC	Crayfish	Cambaridae	GC2	6/17/2014	5.11	-24.94	1	72.41	1.36	2.36	100.07
ER2	ES	GC	Crayfish	Cambaridae	GC2	6/17/2014	6.82	-23.87	1	109.68	0.74	2.36	118.61
ER2	ES	GC	Crayfish	Cambaridae	GC2	6/17/2014	5.21	-25.76	1	156.43	0.21	2.36	212.75
ER2	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/17/2014	1.79	-27.73	13	17.01	1.73	2.36	51.08
ER2	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/17/2014	1.79	-27.67	12	18.46	1.41	2.36	55.45
ER2	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/17/2014	1.3	-27.37	12	15.75	1.36	2.36	57.21
ER2	LS	GC	Crayfish	Cambaridae	GC2	7/22/2014	2.56	-25.23	1	111.34	3.25	2.36	262.88
ER2	LS	SH	Northern caddisfly	Limnephilidae	SH1	7/22/2014	2.17	-27.33	5	23.07	0.63	2.36	61.08
ER2	ES	FC	Brushlegged mayfly	Isonychia	FC1	6/10/2015	5.51	-27.01	16	---	0.19	2.36	---
ER2	ES	GC	Aquatic sowbug	Asellus aquaticus	GC1	6/10/2015	2.55	-26.85	16	---	0.31	2.36	---
ER2	ES	GC	Crayfish	Cambaridae	GC2	6/10/2015	3.31	-30.72	1	249.01	10.74	2.36	486.57

site code	season	feeding group	Common name	Scientific name	feeding group - taxon code	Sample collection date	N15:N14 (per mil)	C13:C12 (per mil)	Individuals in sample	Methylmercury (ng/g dry weight)	Field weight of sample (g)	N15:14 of base consumer for site (per mil)	Calculated methylmercury in trophic position 2.5 consumer (ng/g dry weight)
ER2	ES	GC	Crayfish	Cambaridae	GC2	6/10/2015	4.33	-25.85	3	155.38	5.53	2.36	245.94
ER2	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/10/2015	1.78	-26.86	18	0.90	1.78	2.36	2.72
ER2	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/10/2015	1.23	-27.08	18	17.11	2.32	2.36	64.08
ER2	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/10/2015	2.94	-25.75	17	11.53	1.99	2.36	24.63
ER2	ES	SH	Mortarjoint casemaker	Odontoceridae	SH2	6/10/2015	4.11	-25.42	2	---	0.19	2.36	---
ER2	ES	SH	Shredding stonefly	Plecoptera	SH2	6/10/2015	2.57	-26.75	31	---	0.07	2.36	---
ER2	LS	GC	Aquatic sowbug	Asellus aquaticus	GC1	7/23/2015	4.03	-24.9	5	---	0.12	2.36	---
ER2	LS	GC	Crayfish	Cambaridae	GC2	7/23/2015	5.3	-25.99	1	136.24	0.47	2.36	182.66
ER2	LS	GC	Crayfish	Cambaridae	GC2	7/23/2015	5.07	-25.13	1	170.69	0.72	2.36	237.45
ER2	LS	SH	Northern caddisfly	Limnephilidae	SH1	7/23/2015	1.52	-27.15	9	18.62	0.28	2.36	61.83
ER2	LS	SH	Northern caddisfly	Limnephilidae	SH1	7/23/2015	2.7	-27.87	9	27.57	0.26	2.36	62.67
ER2	LS	SH	Northern caddisfly	Limnephilidae	SH1	7/23/2015	2.29	-27.46	8	30.89	0.29	2.36	78.85
ER2	ES	FC	Brushlegged mayfly	Isonychia	FC1	6/1/2016	3.36	-31.53	41	34.43	0.56	2.36	66.51
ER2	ES	GC	Aquatic sowbug	Asellus aquaticus	GC1	6/1/2016	4.65	-26.92	12	87.39	0.34	2.36	130.54
ER2	ES	GC	Crayfish	Cambaridae	GC2	6/1/2016	4.92	-26.28	1	30.04	3.05	2.36	42.84
ER2	ES	GC	Crayfish	Cambaridae	GC2	6/1/2016	6.13	-25.67	5	182.42	1.64	2.36	216.26
ER2	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/1/2016	2.15	-28.47	6	11.33	0.67	2.36	30.19
ER2	ES	SH	Northern caddisfly	Limnephilidae	SH1	6/1/2016	1.96	-28.52	6	9.70	0.56	2.36	27.49
ER2	ES	SH	Shredding stonefly	Plecoptera	SH2	6/1/2016	3.88	-29.77	13	---	0.04	2.36	---
ER2	LS	FC	Brushlegged mayfly	Isonychia	FC1	7/26/2016	2.98	-30.82	6	---	0.02	2.36	---

site code	season	feeding group	Common name	Scientific name	feeding group - taxon code	Sample collection date	N15:N14 (per mil)	C13:C12 (per mil)	Individuals in sample	Methylmercury (ng/g dry weight)	Field weight of sample (g)	N15:14 of base consumer for site (per mil)	Calculated methylmercury in trophic position 2.5 consumer (ng/g dry weight)
ER2	LS	GC	Aquatic sowbug	Asellus aquaticus	GC1	7/26/2016	4.25	-27.61	71	19.49	0.94	2.36	31.31
ER2	LS	GC	Crayfish	Cambaridae	GC2	7/26/2016	5.43	-27.09	1	53.85	1.8	2.36	70.74
ER2	LS	GC	Crayfish	Cambaridae	GC2	7/26/2016	2.64	-24.45	1	119.50	2.62	2.36	276.02
ER2	LS	GC	Crayfish	Cambaridae	GC2	7/26/2016	5.84	-25.94	2	204.62	1.11	2.36	252.80
ER2	LS	PR	Darner dragonfly	Aeshnidae	PR1	7/26/2016	5.54	-27.98	1	140.75	1.29	2.36	181.82
ER2	LS	SH	Northern caddisfly	Limnephilidae	SH1	7/26/2016	2.35	-29.01	16	23.02	0.59	2.36	57.73

Table 3: The beginning date of each time period. Nutrient sampling of these tributaries began in 2011, however mercury sampling did not commence until 2012. Channel additions occurred approximately annually, while the watershed addition occurred after leaf-off in autumn. EL1 also received one additional calcium carbonate treatment in 2016. All sample collections ended in November of 2016.

Watershed	1	2	3	4	5	6
CL1	Sept 2011	1 Oct 2013	1 March 2014	NA	NA	NA
EL1	Sept 2011	12 July 2012	19 June 2013	28 Feb 2014	16 June 2015	21 June 2016
EL2	Sept 2011	12 July 2012	19 June 2013	28 Feb 2014	16 June 2015	NA

Table 4: Quality assurance/quality control information from COIL and EaSSIL.

Sample ID	Weight (mg)	N2 Amp	%N	δ15N vs. At. Air	CO2 Amp	%C	δ13C vs. VPDB
std CBT	0.943	2,365	12.24	17.76	2,865	48.27	-25.65
std CBT	1.017	2,572	12.28	17.75	3,090	48.45	-25.72
std CBT	1.049	2,661	12.32	17.71	3,183	48.45	-25.65
std CBT	1.015	2,623	12.79	17.28	3,105	48.41	-25.64
std CBT	1.110	2,935	13.03	17.49	3,459	49.59	-25.64
std CBT	0.978	2,520	12.72	17.26	3,002	48.22	-25.66
std KCRN	3.018	1,852	3.00	1.46	7,404	43.15	-13.11
std KCRN	2.991	1,820	3.00	1.54	7,313	43.15	-13.11
std KCRN	3.022	1,828	2.98	1.42	7,347	42.89	-13.12
std KCRN	3.087	1,991	3.17	1.10	7,585	43.07	-13.14
std KCRN	2.99	1,924	3.15	1.07	7,396	42.94	-13.11
std KCRN	2.941	1,800	3.03	1.37	7,266	42.90	-13.13
std Deer	1.07	3,062	13.87	6.40	3,203	47.78	-20.11
std Deer	0.997	2,889	14.17	6.35	3,006	48.17	-20.12
std Deer	1.03	2,967	14.09	6.35	3,069	47.86	-20.19
std Deer	0.961	2,898	14.91	6.28	3,000	50.05	-20.20
std Deer	0.959	2,890	14.84	6.27	2,987	49.69	-20.09
std Deer	1.108	3,230	14.25	6.26	3,305	47.69	-20.13
std Deer	1.082	3,069	13.94	6.25	3,156	46.55	-20.15
std Deer	0.966	2,758	14.17	6.25	2,867	47.22	-20.13

Sample ID	Weight (mg)	N2 Amp	%N	δ15N vs. At. Air	CO2 Amp	%C	δ13C vs. VPDB
std Deer	1.099	3,137	14.03	6.22	3,238	47.04	-20.07
std Deer	1.111	3,214	14.21	6.27	3,314	47.38	-20.17
std methionine	1.07	2,185	10.07	-1.41	2,974	44.07	-25.33
std methionine	0.048	91	10.85	-1.76	137	43.95	-25.12
std methionine	0.112	203	9.56	-1.44	292	40.27	-25.40
std methionine	0.213	362	8.71	-1.81	521	37.88	-25.54
std methionine	0.309	554	9.05	-1.36	785	39.40	-25.37
std methionine	0.514	945	9.16	-1.25	1,323	40.07	-25.32
std methionine	3.049	6,140	9.49	-1.17	7,299	41.58	-25.23
8573	0.481	1042	9.4	-4.7	1067	39.8	-25.8
8573	0.600	1563	5.6	-4.5	1156	21.3	-25.9
8573	0.727	1368	8.2	-4.6	1466	38.4	-26.0
8573	0.305	658	10.2	-4.6	714	45.7	-26.1
8573	0.432	849	9.5	-4.8	935	40.9	-25.9
8573	0.601	1343	10.5	-4.7	1471	45.5	-26.1
8573	0.377	709	9.5	-4.7	786	40.9	-25.9

Sample ID	Weight (mg)	N2 Amp	%N	δ15N vs. At. Air	CO2 Amp	%C	δ13C vs. VPDB
8573	0.385	830	10.9	-4.8	915	46.0	-26.1
Acet	0.788	1761	11.2	-1.1	3075	77.2	-33.2
Acet	0.317	732	11.7	-1.3	1294	79.6	-33.5
Acet	0.547	1295	11.9	-1.1	2268	81.0	-33.4
Acet	0.238	481	9.3	-1.4	853	68.9	-33.3
Acet	0.245	497	10.4	-1.2	880	71.1	-33.3
Acet	0.66	1362	10.4	-1.0	2371	71.1	-33.3
Acet	0.965	1997	10.4	-1.0	3427	71.1	-33.2
Acet	0.484	992	10.4	-1.1	1746	71.7	-33.3
Acet	0.188	450	9.5	-0.9	752	63.1	-33.9
Acet	0.192	468	10.4	-1.4	779	71.1	-33.7
Acet	0.506	1211	10.4	-1.0	2019	71.1	-33.6
Acet	0.885	2143	10.4	-1.0	3443	71.1	-33.5
Acet	0.408	929	9.9	-1.1	1520	67.3	-33.3
Acet	0.917	2175	10.0	-1.0	3476	67.6	-33.2
Acet	0.876	2462	6.4	-0.9	2884	39.3	-33.0
Acet	0.316	653	10.3	-0.9	1165	74.9	-34.2
Acet	0.194	395	10.4	-1.0	679	71.1	-33.2
Acet	0.372	765	9.9	-1.3	1314	68.1	-33.5
Acet	0.695	1518	9.2	-1.0	2573	63.5	-33.3
Acet	0.271	614	10.9	-1.2	1076	75.6	-33.4

Sample ID	Weight (mg)	N2 Amp	%N	δ15N vs. At. Air	CO2 Amp	%C	δ13C vs. VPDB
Acet	0.472	1107	10.3	-0.9	1908	71.8	-33.4
Acet	0.863	2055	9.0	-1.1	3488	62.3	-33.3
Acet	0.166	351	10.5	-1.0	614	72.8	-33.1
Acet	0.198	424	10.4	-1.3	744	71.1	-33.3
Acet	0.389	832	10.4	-1.1	1455	71.1	-33.3
Acet	0.792	1729	10.4	-1.1	2974	71.1	-33.2
Acet	0.560	1204	10.3	-1.3	2098	71.2	-33.4
Acet	0.777	1717	10.4	-1.0	2970	71.9	-33.3
Acet	0.305	707	11.1	-1.3	1244	76.5	-33.5
Acet	0.420	994	11.4	-1.2	1744	78.1	-33.5
Acet	0.865	2138	11.5	-1.0	3672	78.6	-33.3
Acet	0.264	569	10.4	-1.1	929	71.1	-33.3
Acet	0.497	1090	10.4	-1.0	1820	71.1	-33.3
Acet	0.759	1667	10.4	-1.0	2750	71.1	-33.3
Acet	0.623	1359	10.3	-1.2	2264	71.4	-33.2
Acet	0.753	1657	10.4	-1.1	2770	71.9	-33.3
Acet	0.272	628	10.8	-1.2	1066	75.9	-33.6
Acet	0.584	1391	11.1	-1.1	2384	78.0	-33.5
Acet	0.936	2268	11.5	-1.1	3815	78.9	-33.3
Daphnia	0.247	609	10.3	17.3	556	38.9	-24.4
Daphnia	0.523	1125	11.3	17.8	1068	43.7	-24.6

Sample ID	Weight (mg)	N2 Amp	%N	δ15N vs. At. Air	CO2 Amp	%C	δ13C vs. VPDB
Daphnia	0.383	864	10.6	17.8	829	42.8	-24.6
Daphnia	0.409	985	10.9	17.4	952	45.0	-24.6
Daphnia	0.411	933	10.9	17.4	915	42.2	-24.6
Daphnia	0.673	1703	11.8	17.7	1657	45.6	-24.7
Daphnia	0.368	843	10.7	17.2	796	41.8	-24.6
Daphnia	0.460	1144	11.6	17.4	1094	45.4	-24.8
Daphnia	0.687	1601	11.7	17.7	1568	44.9	-24.6
Daphnia	0.724	1783	12.4	17.7	1744	47.2	-24.7
Valine	0.365	1185	7.0	-6.6	883	26.5	-10.4
Valine	0.555	1318	12.3	-6.6	1411	53.9	-10.6
Valine	0.254	663	12.4	-6.6	725	55.9	-10.7
Valine	0.527	1433	11.4	-6.6	1553	53.8	-10.8
Valine	0.538	1451	12.8	-6.6	1595	55.2	-10.7
Valine	0.684	1944	13.3	-6.7	2120	57.3	-10.7
Valine	0.415	1110	12.6	-6.7	1178	54.9	-10.7
Valine	0.221	611	12.9	-6.7	657	56.6	-10.8
Valine	0.381	1014	13.5	-6.7	1121	57.1	-10.71

Table 5: Regression Adj-R^2, slopes and p-values for relationship between mercury species and other analytes.

			Source									
			CL1	**EL1**	**EL2**	**EL1P**	**ER1**	**EL1M**	**CR1**	**ER2**	**EL1R**	**EL2R**
MeHg	**Dissolved Organic Carbon**	**Adj-R^2**	-0.024	**0.2171**	0.0093	**0.3322**	0.0713	-0.0042	-0.0463	-0.0698	-0.0074	-0.0178
		p-value	0.5042	**0.0021**	0.2776	**0.0182**	0.1342	0.3595	0.8746	0.5222	0.3884	0.436
		Slope	0.0051	**0.1734**	0.0476	**0.0384**	0.1005	-0.0574	0.0024	0.0471	0.0548	-0.0156
	Nitrate	**Adj-R^2**	0.0035	-0.0053	0.0502	**0.3046**	0.0985	**0.1709**	0.0626	0.9723	-0.0303	-0.0342
		p-value	0.3097	0.374	0.1408	**0.0238**	0.0965	**0.0097**	0.1311	0.0751	0.8106	0.5869
		Slope	-0.0008	-0.0208	-0.0059	**-0.1487**	-0.0565	**0.0345**	-0.0011	-0.0086	0.0031	0.0009
	Organic Monomeric Aluminum	**Adj-R^2**	0.0195	**0.4162**	-0.0431	0.0205	-0.0412	**0.1224**	0.0687	-0.3367	-0.0027	-0.0445
		p-value	0.2437	**<0.0001**	0.9277	0.2872	0.5987	**0.0281**	0.1261	0.6093	0.3463	0.7494
		Slope	-0.0152	**0.5039**	-0.0051	0.3974	-0.0368	**0.1718**	-0.0185	-0.1322	-0.1012	-0.0059
	pH	**Adj-R^2**	-0.0454	0.0317	-0.0411	-0.0753	**0.1694**	-0.0235	0.1	0.9198	-0.0303	-0.0442
		p-value	0.9808	0.149	0.9078	0.7694	**0.0405**	0.6094	0.0775	0.1283	0.8086	0.7434
		Slope	-0.0006	-0.4072	0.0129	0.1033	**0.6572**	-0.0673	0.3689	0.2649	0.1041	-0.0142
	Specific Conductance	**Adj-R^2**	-0.0218	-0.0263	-0.0114	0.0787	0.0931	0.0075	**0.2654**	0.5006	0.0299	-0.027
		p-value	0.4827	0.7826	0.4049	0.172	0.103	0.2738	**0.007**	0.3331	0.1685	0.5109
		Slope	-0.0015	0.0038	-0.0031	0.0041	-0.0902	-0.0042	**-0.0167**	0.0285	-0.0501	0.0034
	Sulfate	**Adj-R^2**	-0.0077	0.076	0.0011	-0.0817	0.0928	0.0084	0.0236	**0.9955**	0.0021	-0.0433
		p-value	0.374	0.0544	0.3206	0.8947	0.1034	0.2682	0.2294	**0.0301**	0.3095	0.724
		Slope	-0.1037	-1.4223	-0.1813	-0.157	-1.1165	0.4644	-0.1426	**1.5003**	-0.4632	0.0491
	SUVA	**Adj-R^2**	-0.0453	0.0704	-0.0407	**0.2616**	0.0232	**0.3472**	-0.0433	NaN	-0.0322	0.0584
		p-value	0.9616	0.0645	0.8069	**0.0356**	0.2485	**0.0002**	0.7697	NaN	0.8564	0.1508
		Slope	0.4915	63.6482	-4.6568	**-30.876**	31.6136	**102.2319**	1.4756	-19050.9179	3.8774	-16.7955
	THg	**Adj-R^2**	-0.0354	**0.5898**	0.1051	**0.5519**	-0.0133	-0.026	-0.0434	-0.4825	**0.297**	-0.0251
		p-value	0.6486	**<0.0001**	0.0588	**0.0022**	0.3979	0.6874	0.7725	0.6603	**0.0007**	0.4935
		Slope	0.0064	**0.3677**	0.1693	**0.4863**	0.0962	0.0574	-0.0073	0.1504	**0.3186**	0.0268

			Source									
			CL1	EL1	EL2	EL1P	ER1	EL1M	CR1	ER2	EL1R	EL2R
	Total Monomeric Aluminum	Adj-R^2	0.0538	**0.2795**	-0.0393	0.198	0.0206	0.0384	**0.1514**	-0.0517	0.0044	-0.05
		p-value	0.1484	**0.0005**	0.763	0.0719	0.2566	0.145	**0.0415**	0.5165	0.2945	0.9923
		Slope	-0.0099	**0.3363**	-0.0142	0.5369	-0.052	0.0838	**-0.0088**	-0.0851	-0.0665	-0.0001
	UV254	Adj-R^2	-0.0274	**0.2713**	-0.0065	**0.3934**	0.0739	-0.03	-0.0473	NaN	0.0125	0.0147
		p-value	0.5404	**0.0007**	0.3676	**0.0097**	0.1369	0.7963	0.9347	NaN	0.2471	0.2686
		Slope	0.1035	**3.5526**	1.0134	**3.5183**	1.682	0.3952	0.0305	0.6707	2.0788	-0.5285
THg	Dissolved Organic Carbon	Adj-R^2	**0.83**	**0.263**	**0.4644**	0.1107	**0.2556**	**0.1519**	**0.63**	-0.2872	**0.1918**	0.029
		p-value	**<0.0001**	**0.0006**	**<0.0001**	0.1426	**0.0081**	**0.0143**	**<0.0001**	0.6233	**0.0071**	0.2168
		Slope	**0.4997**	**0.3966**	**0.3304**	0.0386	**0.2842**	**0.1834**	**0.4885**	0.2579	**0.287**	0.1405
	MeHg	Adj-R^2	-0.0354	**0.5898**	0.1051	**0.5519**	-0.0133	-0.026	-0.0434	-0.4825	**0.297**	-0.0251
		p-value	0.6486	**<0.0001**	0.0588	**0.0022**	0.3979	0.6874	0.7725	0.6603	**0.0007**	0.4935
		Slope	1.4922	**1.6349**	0.8323	**1.2117**	0.4159	0.0894	-0.5584	1.7202	**1.0036**	0.8871
	Nitrate	Adj-R^2	-0.0298	0.0556	**0.1521**	**0.2936**	**0.1413**	0.0466	0.1001	0.2572	0.09	-0.0383
		p-value	0.6034	0.0831	**0.0209**	**0.0324**	**0.0434**	0.1194	0.0775	0.2895	0.0528	0.6396
		Slope	-0.0062	-0.0829	**-0.0168**	**-0.2186**	**-0.043**	-0.0268	-0.0114	-0.0526	-0.0431	-0.0043
	Organic Monomeric Aluminum	Adj-R^2	**0.1711**	**0.4236**	**0.3294**	-0.0383	**0.3467**	-0.033	**0.6221**	-0.4929	-0.0326	0.0945
		p-value	**0.0228**	**<0.0001**	**0.0008**	0.4585	**0.0023**	0.9226	**<0.0001**	0.9313	0.8175	0.0892
		Slope	**0.4654**	**1.0582**	**0.3454**	0.3905	**0.3373**	0.0097	**0.4234**	0.1654	0.0443	0.1756
	pH	Adj-R^2	**0.2055**	0.0228	0.0399	-0.0909	-0.0446	-0.001	-0.0431	-0.4963	-0.0312	-0.0474
		p-value	**0.0116**	0.1806	0.1528	0.9948	0.8087	0.3329	0.7669	0.9502	0.8042	0.8256
		Slope	**0.8688**	-0.7825	-0.3546	0.0036	-0.156	0.1579	0.5627	0.1498	-0.1888	-0.0549
	Specific Conductance	Adj-R^2	**0.2556**	-0.0238	0.0614	0.0207	-0.0425	-0.0244	0.0336	-0.3513	0.0888	-0.0399
		p-value	**0.0049**	0.7123	0.1039	0.2866	0.7523	0.6286	0.1981	0.6851	0.054	0.6642
		Slope	**0.0829**	-0.0104	-0.0133	0.005	-0.0304	-0.0023	-0.0744	0.1164	-0.1246	0.0129
	Sulfate	Adj-R^2	0.1126	**0.1652**	0.0422	-0.0783	**0.1336**	0.0409	0.1128	-0.2823	0.0616	-0.0379

		Source									
		CL1	**EL1**	**EL2**	**EL1P**	**ER1**	**EL1M**	**CR1**	**ER2**	**EL1R**	**EL2R**
	p-value	0.0523	**0.0066**	0.1467	0.7271	**0.0484**	0.1342	0.0648	0.619	0.0917	0.6343
	Slope	-3.197	**-4.0753**	-0.5999	-0.6975	**-2.1363**	-0.776	-1.8738	-5.4872	-1.3482	-0.38
SUVA	**Adj-R^2**	**0.3751**	**0.1504**	-0.0368	0.214	-0.012	0.0298	-0.0176	0.8511	0.0485	-0.0401
	p-value	**0.0005**	**0.0102**	0.8415	0.0633	0.3967	0.1691	0.44	0.1759	0.1227	0.6381
	Slope	**448.472**	**177.2647**	8.5292	-42.0602	46.0738	-51.4858	33.8801	-51081.8144	-56.9817	-32.553
Total Monomeric Aluminum	**Adj-R^2**	-0.0417	**0.2703**	**0.3254**	0.1168	**0.1867**	-0.0324	-0.0241	-0.4891	-0.0325	0.0973
	p-value	0.8462	**0.0006**	**0.0009**	0.1483	**0.0256**	0.8681	0.4849	0.9146	0.8153	0.0858
	Slope	0.0222	**0.6924**	**0.2928**	0.6103	**0.1745**	-0.0119	0.0302	-0.1162	-0.0275	0.1221
UV254	**Adj-R^2**	**0.8453**	**0.3834**	**0.483**	0.1878	**0.1651**	**0.1031**	**0.5194**	-0.6991	**0.1685**	0.0222
	p-value	**<0.0001**	**<0.0001**	**<0.0001**	0.0781	**0.0344**	**0.0384**	**0.0001**	0.7464	**0.0126**	0.2426
	Slope	**11.1151**	**8.7142**	**8.2278**	3.8762	**4.0627**	**3.8161**	**10.8515**	5.8342	**7.573**	3.2183

Table 6: Summary statistics for each site are divided into their respective treatment periods

CL1

		1		2		3	
Site	**Variable**	**mean**	**SD**	**mean**	**SD**	**mean**	**SD**
CL1	DOC	5.510	1.641	10.553	4.065	8.290	3.049
CL1	Silicon	74.885	18.388	90.534	11.163	82.678	9.995
CL1	MeHg	0.026	0.019	0.074	0.065	0.101	0.099
CL1	NO_3	1.452	1.315	2.466	1.268	1.601	1.031
CL1	pH	4.540	0.089	6.599	0.607	5.183	0.175
CL1	SO_4-S	0.991	0.164	1.171	0.247	0.958	0.183
CL1	SUVA	0.039	0.003	0.042	0.002	0.042	0.002
CL1	THg	2.058	0.919	3.266	1.731	2.329	0.944
CL1	UV_{254}	0.233	0.068	0.451	0.179	0.347	0.132
CR1	DOC	4.486	1.019	5.434	1.342	4.673	1.272
CR1	Silicon	60.357	10.314	57.440	4.037	56.755	9.621
CR1	MeHg	0.002	0.003	0.019	0.015	0.073	0.069
CR1	NO_3	1.312	1.133	1.550	1.070	1.148	0.895
CR1	pH	4.600	0.085	4.584	0.055	4.601	0.073
CR1	SO_4-S	0.927	0.149	0.824	0.092	0.785	0.118
CR1	SUVA	0.034	0.003	0.035	0.002	0.035	0.002
CR1	THg	1.540	0.535	1.367	0.613	0.905	0.366
CR1	UV_{254}	0.161	0.043	0.191	0.052	0.162	0.045

EL1

		1		2		3		4		5		6	
Site	**Variable**	**mean**	**SD**	**mean**	**SD**	**mean**	**SD**	**mean**	**SD**	**mean**	**SD**	**mean**	**SD**
EL1	DOC	7.550	2.817	8.758	2.586	9.283	1.677	7.258	1.571	7.783	1.582	11.625	2.275
EL1	Silicon	55.022	23.446	61.422	20.588	51.201	14.236	45.190	29.224	46.988	17.106	42.723	15.235
EL1	MeHg	2.600	3.111	1.222	0.880	0.498	0.432	0.260	0.175	0.514	0.139	0.928	0.821
EL1	NO_3	0.923	1.053	0.604	0.522	0.599	0.572	0.680	0.562	0.411	0.292	0.258	0.187
EL1	pH	5.162	0.263	6.029	0.485	6.285	0.579	5.880	0.395	6.093	0.508	6.322	0.485
EL1	SO_4-S	0.707	0.200	0.716	0.297	0.584	0.147	0.615	0.189	0.623	0.170	0.392	0.075
EL1	SUVA	0.050	0.006	0.050	0.007	0.047	0.003	0.046	0.002	0.047	0.003	0.050	0.006
EL1	THg	6.170	5.770	4.610	1.061	2.127	1.410	1.360	0.495	1.805	0.459	3.485	1.339
EL1	UV_{254}	0.491	0.169	0.450	0.189	0.436	0.085	0.337	0.069	0.364	0.087	0.579	0.090
EL1M	DOC	NA	NA	9.448	5.507	8.461	1.386	6.972	1.340	7.601	1.553	9.179	0.714
EL1M	Silicon	NA	NA	35.127	24.701	25.977	13.976	43.353	36.946	26.883	21.070	24.598	15.994
EL1M	MeHg	NA	NA	0.476	NA	0.601	0.468	0.739	0.708	0.378	0.225	0.655	0.424
EL1M	NO_3	NA	NA	0.301	0.556	0.273	0.578	0.531	0.694	0.169	0.293	0.042	0.025
EL1M	pH	NA	NA	6.245	0.767	6.799	0.789	5.536	0.456	6.001	0.709	6.619	0.704
EL1M	SO_4-S	NA	NA	0.529	0.229	0.523	0.127	0.580	0.200	0.553	0.186	0.361	0.084

		1		2		3		4		5		6	
Site	**Variable**	**mean**	**SD**	**mean**	**SD**	**mean**	**SD**	**mean**	**SD**	**mean**	**SD**	**mean**	**SD**
EL1M	SUVA	NA	NA	0.041	0.006	0.045	0.002	0.045	0.004	0.043	0.002	0.043	0.002
EL1M	THg	NA	NA	1.780	NA	1.775	0.816	1.255	0.337	1.406	0.275	2.010	0.661
EL1M	UV_{254}	NA	NA	0.359	0.134	0.378	0.064	0.319	0.047	0.329	0.061	0.395	0.035
EL1P	MeHg	NA	NA	NA	NA	NA	NA	NA	NA	0.152	0.059	0.615	0.434
EL1P	THg	NA	NA	NA	NA	NA	NA	NA	NA	1.225	0.431	1.874	0.648
EL1R	DOC	8.194	2.009	8.161	1.801	8.795	1.882	6.867	1.369	7.687	1.745	8.601	0.733
EL1R	Silicon	16.166	7.867	35.342	25.101	29.837	18.752	44.583	40.133	31.468	21.049	23.978	17.111
EL1R	MeHg	1.240	1.131	0.416	0.329	0.411	0.433	0.554	0.315	0.291	0.176	0.720	0.367
EL1R	NO_3	0.133	0.217	0.259	0.521	0.416	0.805	0.588	0.726	0.231	0.384	0.029	0.023
EL1R	pH	5.066	0.142	5.123	0.300	4.931	0.149	4.953	0.285	4.971	0.317	5.020	0.099
EL1R	SO_4-S	0.547	0.163	0.557	0.225	0.533	0.170	0.627	0.199	0.518	0.160	0.357	0.087
EL1R	SUVA	0.040	0.005	0.042	0.004	0.045	0.002	0.046	0.004	0.043	0.003	0.044	0.004
EL1R	THg	3.215	1.775	2.080	0.180	1.677	1.012	1.080	0.349	1.254	0.305	1.790	0.390
EL1R	UV_{254}	0.375	0.083	0.335	0.056	0.396	0.087	0.322	0.050	0.330	0.066	0.376	0.028
ER1	DOC	NA	NA	5.955	2.349	7.461	2.358	6.214	1.561	7.401	1.078	7.095	0.464

		1		2		3		4		5		6	
Site	**Variable**	**mean**	**SD**	**mean**	**SD**	**mean**	**SD**	**mean**	**SD**	**mean**	**SD**	**mean**	**SD**
ER1	Silicon	NA	NA	52.665	6.731	65.517	18.444	45.412	15.084	45.343	22.491	66.925	25.407
ER1	MeHg	NA	NA	0.474	NA	0.568	0.380	0.389	0.498	0.443	0.378	0.596	0.318
ER1	NO_3	NA	NA	0.260	0.238	0.403	0.916	0.461	0.786	0.014	0.014	0.047	0.061
ER1	pH	NA	NA	4.901	0.111	5.055	0.302	4.879	0.180	5.203	0.264	5.340	0.137
ER1	SO_4-S	NA	NA	0.875	0.190	0.726	0.237	0.744	0.144	0.728	0.108	0.754	0.045
ER1	SUVA	NA	NA	0.046	0.000	0.047	0.004	0.045	0.003	0.046	0.003	0.044	0.003
ER1	THg	NA	NA	2.390	NA	2.218	1.170	1.247	0.219	1.213	0.320	2.460	0.469
ER1	UV_{254}	NA	NA	0.272	0.105	0.354	0.127	0.297	0.078	0.347	0.075	0.313	0.028
ER2	DOC	5.467	2.529	7.707	4.318	8.792	5.140	8.420	5.034	7.192	4.981	7.740	3.081
ER2	Silicon	133.483	58.453	165.847	58.139	131.048	43.994	114.299	43.050	173.512	56.562	164.629	24.672
ER2	MeHg	0.155	0.106	0.340	NA	NaN	NA	NA	NA	NA	NA	NA	NA
ER2	NO_3	2.078	1.075	1.151	0.843	0.986	0.616	1.043	0.775	0.738	0.502	0.459	0.293
ER2	pH	5.644	0.613	6.302	0.487	5.999	0.591	5.702	0.601	6.092	0.534	6.189	0.341
ER2	SO_4-S	1.072	0.172	1.105	0.252	0.978	0.199	0.931	0.284	1.133	0.181	1.014	0.105
ER2	SUVA	0.045	0.002	0.045	0.003	0.045	0.002	0.067	0.098	0.044	0.002	0.044	0.002

		1		2		3		4		5		6	
Site	**Variable**	**mean**	**SD**	**mean**	**SD**	**mean**	**SD**	**mean**	**SD**	**mean**	**SD**	**mean**	**SD**
ER2	THg	2.620	0.622	2.560	NA	0.310	NA	NA	NA	NA	NA	NA	NA
ER2	UV_{254}	0.287	0.187	0.339	0.176	0.390	0.213	0.472	0.418	0.312	0.208	0.338	0.121

EL2

		1		2		3		4		5	
Site	**Variable**	**mean**	**SD**	**mean**	**SD**	**mean**	**SD**	**mean**	**SD**	**mean**	**SD**
EL2	DOC	4.386	1.008	5.251	1.886	6.166	1.814	5.286	2.382	5.732	2.640
EL2	Silicon	130.846	54.275	141.608	56.822	99.330	39.672	96.597	37.633	109.405	48.956
EL2	MeHg	0.030	0.042	0.106	0.008	0.466	0.740	0.043	0.028	0.155	0.213
EL2	NO_3	2.292	1.303	1.543	0.848	0.933	0.798	1.585	1.169	0.823	0.452
EL2	pH	5.577	0.531	7.102	0.574	7.205	0.538	6.635	0.402	6.822	0.345
EL2	SO_4-S	1.032	0.177	1.176	0.262	0.904	0.157	0.870	0.237	0.961	0.333
EL2	SUVA	0.043	0.004	0.042	0.003	0.041	0.002	0.041	0.003	0.041	0.002
EL2	THg	1.610	0.255	2.330	0.594	1.830	0.271	0.837	0.478	1.180	0.566
EL2	UV_{254}	0.205	0.047	0.215	0.075	0.256	0.079	0.217	0.100	0.233	0.109
EL2R	DOC	3.670	0.588	4.567	1.702	4.937	0.680	4.446	1.545	4.912	1.204

		1		2		3		4		5	
Site	**Variable**	**mean**	**SD**	**mean**	**SD**	**mean**	**SD**	**mean**	**SD**	**mean**	**SD**
EL2R	Silicon	158.881	95.206	150.391	66.503	119.458	39.035	97.598	36.385	120.793	51.660
EL2R	MeHg	0.120	0.099	0.061	0.055	0.046	0.037	0.116	0.166	0.102	0.035
EL2R	NO_3	2.015	0.685	1.460	0.987	1.110	0.943	1.653	1.225	0.717	0.407
EL2R	pH	5.951	0.804	5.966	0.636	5.724	0.557	5.500	0.560	5.662	0.412
EL2R	SO_4-S	1.159	0.211	1.189	0.213	0.997	0.089	0.983	0.172	0.987	0.124
EL2R	SUVA	0.038	0.002	0.043	0.004	0.042	0.002	0.041	0.002	0.042	0.002
EL2R	THg	1.660	0.212	2.210	0.438	2.123	0.395	1.050	0.510	1.037	0.256
EL2R	UV_{254}	0.143	0.021	0.194	0.069	0.208	0.031	0.187	0.067	0.203	0.048
ER1	DOC	NA	NA	5.955	2.349	8.324	1.171	5.873	1.782	7.325	0.950
ER1	Silicon	NA	NA	52.665	6.731	59.272	11.517	51.116	22.178	50.739	24.094
ER1	MeHg	NA	NA	0.474	NA	0.568	0.380	0.389	0.498	0.489	0.351
ER1	NO_3	NA	NA	0.260	0.238	0.030	0.022	0.662	0.951	0.022	0.032
ER1	pH	NA	NA	4.901	0.111	5.054	0.337	4.899	0.179	5.238	0.241
ER1	SO_4-S	NA	NA	0.875	0.190	0.637	0.102	0.792	0.197	0.735	0.095
ER1	SUVA	NA	NA	0.046	0.000	0.048	0.003	0.045	0.003	0.045	0.003

		1		2		3		4		5	
Site	**Variable**	**mean**	**SD**	**mean**	**SD**	**mean**	**SD**	**mean**	**SD**	**mean**	**SD**
ER1	THg	NA	NA	2.390	NA	2.712	0.442	1.079	0.455	1.525	0.658
ER1	UV_{254}	NA	NA	0.272	0.105	0.399	0.071	0.276	0.093	0.338	0.066
ER2	DOC	5.467	2.529	7.707	4.318	9.079	5.099	8.165	5.044	7.452	4.087
ER2	Silicon	133.483	58.453	165.847	58.139	130.612	45.090	115.512	42.326	169.304	43.485
ER2	MeHg	0.155	0.106	0.340	NA	NA	NA	NaN	NA	NA	NA
ER2	NO_3	2.078	1.075	1.151	0.843	0.888	0.434	1.133	0.861	0.606	0.430
ER2	pH	5.644	0.613	6.302	0.487	5.978	0.599	5.735	0.606	6.138	0.443
ER2	SO_4-S	1.072	0.172	1.105	0.252	0.963	0.191	0.948	0.287	1.077	0.158
ER2	SUVA	0.045	0.002	0.045	0.003	0.045	0.002	0.066	0.096	0.044	0.002
ER2	THg	2.620	0.622	2.560	NA	NA	NA	0.310	NA	NA	NA
ER2	UV_{254}	0.287	0.187	0.339	0.176	0.402	0.211	0.456	0.414	0.324	0.168

Table 7: BACI table of the interaction of THg between EL1 and EL1R. Estimates and Standard error are based on the calculated difference between the two sites and periods.

Period	Estimate	Std error	F-statistic	p-value
1-2	0.425	1.507	0.282	0.779
1-3	2.505	1.364	1.837	0.071
1-4	2.675	1.262	2.119	0.038
1-5	2.404	1.221	1.970	0.054
1-6	1.260	1.232	1.023	0.311
2-3	2.080	1.286	1.618	0.111
2-4	2.250	1.178	1.910	0.061
2-5	1.979	1.133	1.747	0.086
2-6	0.835	1.145	0.729	0.469
3-4	0.170	0.987	0.172	0.864
3-5	-0.101	0.933	-0.108	0.914
3-6	-1.245	0.948	-1.313	0.194
4-5	-0.271	0.778	-0.348	0.729

Period	Estimate	Std error	F-statistic	p-value
4-6	-1.415	0.796	-1.778	0.081
5-6	-1.144	0.728	-1.573	0.121

Table 8: Average mass of molecules given particular numbers of sulfur in the condensed and unsaturated hydrocarbon regions of the van Krevelen diagram.

S	Average Mass	Standad Deviation
1	579.7444	97.22022
2	521.6482	135.81923
4	497.1550	102.34782

Table 9: Percent composition of each sample analyzed by ESI-FTICR-MS. The columns are labeled as 'treatment level_period month/day'

Candidates	R_1 9/25	R_2 10/17	R_2 10/22	R_2 10/9	R_3 7/26	R_3 9/19	T_1 9/25	T_2 10/17	T_2 10/22	T_2 10/9	T_2 11/18	T_3 7/26	T_3 8/24
CHO	68.4	68.0	71.7	69.6	73.3	73.9	70.6	57.9	60.8	57.2	73.5	69.3	70.0
CHON	11.4	11.6	14.1	10.1	9.7	8.7	11.3	6.9	7.9	11.2	4.9	8.6	10.4
CHONP	2.7	2.6	1.2	2.8	2.0	2.3	2.0	6.3	5.0	4.4	3.8	3.4	2.6
CHONS	1.7	1.3	1.6	1.2	1.1	1.2	1.7	1.4	1.2	2.5	0.8	1.0	1.4
CHONSP	3.5	3.3	2.4	3.1	2.6	2.7	3.1	5.2	4.8	3.7	3.7	3.5	3.2
CHOP	0.9	0.7	0.2	0.7	1.1	1.0	0.5	1.3	1.3	0.5	0.4	0.5	1.0
CHOS	11.1	12.3	8.7	12.5	10.2	10.2	10.8	21.0	19.0	20.4	12.8	13.7	11.4
CHOSP	0.1	0.0	0.1	0.1	0.0	0.1	0.1	0.1	0.1	0.1	0.0	0.0	0.0

Table 10: Preliminary information on selected samples for analysis by FTICR-MS. These paired (limed – reference) samples were collected before, immediately after, and three years after watershed lime addition. They represent the full range in tributary response to watershed lime addition. The blue, red, and green highlighting correspond to data points in Figure 1.

Source	Date Sampled	pH	MeHg (µmol/L)	THg (µmol/L)	Sp C	DOC (µmol/L)	Chloride (µmol/L)	Sulfate (µmol/L)	UV254 (m^{-})	THg:DOC (mmol:mol)	SUVA
Limed	6/11/2013	4.41	0.037	16.49	21.40	733.0	4.6	23.20	0.372	2.25E-05	0.0423
Limed	9/25/2013	4.62	0.204	10.50	19.81	423.9	6.6	34.44	0.200	2.48E-05	0.0393
Limed	10/9/2013	7.15	0.380	27.23	51.70	1073.2	77.5	41.48	0.559	2.54E-05	0.0434
Limed	10/17/2013	7.00	0.538	20.25	38.60	793.2	40.2	36.68	0.425	2.55E-05	0.0446
Limed	10/22/2013	6.63	0.288	16.58	31.30	739.4	55.0	35.50	0.376	2.24E-05	0.0423
Limed	10/30/2013	6.49	0.195	11.88	25.10	523.3	44.3	35.90	0.259	2.27E-05	0.0412
Limed	7/26/2016	5.19	0.964	14.50	14.84	565.4	6.5	34.64	0.302	2.57E-05	0.0445
Limed	8/24/2016	5.14	1.166	13.91	14.74	643.4	4.9	31.04	0.319	2.16E-05	0.0413
Limed	9/19/2016	5.15	1.214	16.34	15.08	855.7	7.8	25.83	0.412	1.91E-05	0.0401
Reference	6/11/2013	4.50	BDL	10.64	18.67	500.8	5.7	24.41	0.227	2.13E-05	0.0377
Reference	9/25/2013	4.64	0.028	6.53	17.69	363.7	6.5	32.21	0.151	1.80E-05	0.0346
Reference	10/9/2013	4.61	0.037	10.54	18.43	455.9	8.0	28.15	0.184	2.31E-05	0.0336
Reference	10/17/2013	4.65	0.056	7.18	17.89	398.9	7.9	30.23	0.170	1.80E-05	0.0355
Reference	10/22/2013	4.61	0.042	7.72	19.07	413.2	7.7	27.72	0.168	1.87E-05	0.0338
Reference	10/30/2013	4.64	0.158	7.38	18.52	368.5	6.1	28.55	0.150	2.00E-05	0.0339
Reference	7/26/2016	4.61	0.713	4.55	17.32	325.8	9.4	28.88	0.138	1.40E-05	0.0353
Reference	8/24/2016	4.68	0.778	5.74	15.50	400.1	4.0	26.09	0.161	1.44E-05	0.0335
Reference	9/19/2016	4.62	0.788	7.97	15.94	446.8	5.7	23.74	0.168	1.78E-05	0.0313

Table 11: Water chemistry in 35 lakes in 2010s in New York State. N/A indicate values that are below the detection limit.

Location	Lake	ANC (µeq L^{-1})	pH	Chl (µg L^{-1})	DOC (mg L^{-1})	F (mg L^{-1})	Cl (mg L^{-1})	SO4 (mg L^{-1})	NO3 (mg L^{-1})	NH4 (mg L^{-1})	Total Al (µg L^{-1})	MeHg (ng L^{-1})	Total Hg (ng L^{-1})
Northeast	Big Moose Lake	46	5.6	4.6	5.6	0.04	0.2	0.7	N/A	0.08	81.6	0.06	1.12
	Delta Lake	1157	6.9	5.7	2.4	0.04	5.0	1.4	0.14	0.002	52.6	0.04	0.4
	Fall Lake	220	6.3	4.7	10.1	0.10	2.4	0.7	N/A	0.01	61.8	0.38	1.18
	Ferris Lake	23	6.0	5.3	4.0	N/A	0.2	0.7	N/A	0.01	7.8	0.09	0.78
	Francis Lake	0.4	6.4	4.3	4.4	0.09	0.2	0.8	N/A	0.20	2.9	0.04	0.92
	Hinckley Reservoir	230	6.6	4.1	5.2	0.04	2.4	0.9	0.06	0.06	61.7	0.01	0.76
	Kings Flow	77	6.1	5.2	6.0	0.05	0.2	0.7	N/A	0.002	57.0	0.10	0.76
	Limekiln Lake	36	6.0	3.4	3.5	0.04	0.6	0.7	N/A	0.08	0.5	0.03	0.42
	Middle Stoner Lake	112	6.2	4.2	4.3	0.04	15.7	0.9	0.0002	N/A	54.7	0.05	0.51
	North Lake	45	5.5	3.6	6.4	0.06	0.2	0.8	0.16	0.06	91.2	0.06	1.52
	Payne Lake	1043	6.5	43.0	16.7	0.06	0.2	0.7	N/A	0.002	44.1	0.01	0.36
	Red Lake	1100	6.7	14.3	16.7	0.09	15.9	1.5	0.04	0.002	2.7	0.03	N/A

	Rock Pond	68	6.0	2.5	15.8	0.05	0.2	0.5	N/A	0.05	39.6	N/A	2.37
	Sand Lake	23	4.8	3.1	6.4	0.11	0.7	0.7	0.0009	N/A	82.0	0.11	1.55
	Soft Maple Dam Pond	43	6.5	1.8	5.1	0.05	8.2	1.8	0.04	0.09	7.8	0.07	1.18
	Spy Lake	73	6.1	9.3	3.7	0.02	15.6	0.7	0.0003	N/A	52.0	0.04	1.19
	Sunday Lake	58	6.7	2.9	11.1	0.11	0.2	0.6	0.03	0.15	147.4	0.31	3.64
	West Caroga Lake	347	6.7	2.2	4.5	0.03	15.7	1.0	N/A	0.04	54.2	0.04	0.55
Southeast	Canadarago Lake	2546	8.3	3.6	N/A	0.04	N/A	3.3	0.1	0.12	4.7	0.02	0.22
	East Sidney Reservoir	627	8.2	9.9	2.1	0.04	11.8	1.8	0.3	0.22	4.5	N/A	0.4
	Forge Pond	352	6.3	5.5	5.5	N/A	N/A	N/A	N/A	0.00	56.9	0.03	N/A
	Fort Pond	25	6.4	7.4	5.8	0.07	69.0	0.3	3.7	0.20	43.1	0.09	N/A
	Fresh Pond	279	5.6	16.1	5.3	0.03	24.5	1.9	0.0	0.12	60.9	0.06	0.75
	Goodyear Lake	1902	8.2	6.0	3.3	0.05	17.0	2.4	0.3	0.01	4.5	0.02	0.56
	Greenwood Lake	671	9.3	41.9	3.4	0.06	N/A	2.6	N/A	0.63	5.1	0.02	0.24
	Lake Huntington	495	8.8	5.4	5.0	0.05	N/A	1.4	N/A	0.27	-0.2	0.01	0.5
	Lake Superior	176	7.8	5.9	5.7	0.04	13.5	1.3	N/A	0.17	2.6	0.03	0.65
	Mongaup Pond	154	8.6	6.9	2.1	0.03	0.5	1.2	0.1	0.25	2.2	0.03	0.58

	Onteora Lake	267	7.7	29.8	5.2	0.04	1.6	1.2	N/A	0.31	3.8	0.05	0.6
	Otsego Lake	2406	8.3	2.5	2.2	0.04	16.1	3.1	0.5	0.09	-1.0	0.01	N/A
	Rio Reservoir	153	8.4	5.1	3.3	0.04	N/A	1.8	N/A	0.21	1.4	0.02	0.43
	Swinging Bridge Reservoir	265	7.9	9.4	4.0	0.04	N/A	1.7	N/A	0.17	4.4	0.04	0.63
	White Pond	461	9.3	1.5	2.6	0.03	N/A	2.6	N/A	0.09	3.8	0.02	0.27
West	Eaton Brook Reservoir	1454	6.8	1.5	2.2	0.04	5.8	1.4	0.04	0.004	50.6	0.02	N/A
	Lake Moraine	1930	6.7	25.2	2.6	0.04	18.6	1.8	0.06	0.002	7.0	0.07	N/A

Table 12: Correlation coefficients among water chemistry variables and fish THg concentrations. Values (r^2) in bold indicated a *p* value less than 0.05.

	Fish THg concentrations				Surface water chemistry													
	YP	WEYE	SMB	LMB	ANC	pH	chl	DOC	F	Cl	SO4	NO3	NH4	Total Al	Org Al	Inorg Al	MeHg	THg
ANC	**-0.5**	0.2	**-0.6**	-0.03	1	**0.9**	0.1	-0.2	0.03	**0.6**	**0.7**	0.3	0.3	-0.3	**-0.4**	-0.02	-0.3	**-0.4**
pH	**-0.6**	0.08	**-0.6**	-0.4	-	1	0.3	**-0.4**	-0.01	**0.6**	**0.7**	0.2	0.1	**-0.4**	**-0.4**	-0.02	**-0.3**	**-0.5**
Chl	**-0.4**	-0.2	-0.1	**-0.7**	-	-	1	0.3	-0.04	0.02	-0.06	**-0.6**	-0.1	-0.3	**-0.4**	-0.04	0.06	-0.04
DOC	0.2	0.3	0.1	-0.2	-	-	-	1	0.3	-0.3	**-0.5**	-0.4	0.05	0.2	**0.4**	-0.03	**0.8**	**0.7**
F	**0.5**	-0.03	-0.2	-0.2	-	-	-	-	1	-0.4	-0.1	0.3	0.1	**0.4**	**0.4**	0.2	0.2	0.3
Cl	**-0.6**	-0.7	-0.4	**0.9**	-	-	-	-	-	1	**0.7**	-0.1	0.04	**-0.5**	**-0.6**	-0.3	**-0.5**	**-0.5**
SO4	**-0.6**	-0.3	**-0.8**	0.5	-	-	-	-	-	-	1	0.2	0.2	-0.2	**-0.3**	0.1	**-0.7**	**-0.7**
NO3	0.1	0.2	-0.2	-0.4	-	-	-	-	-	-	-	1	0.3	0.3	0.3	0.2	-0.1	-0.001
NH4	0.01	**0.7**	-0.4	0.1	-	-	-	-	-	-	-	-	1	0.08	0.2	-0.2	**0.4**	0.1
Total Al	**0.6**	-0.01	**-0.7**	0.5	-	-	-	-	-	-	-	-	-	1	**0.8**	**0.7**	**0.3**	**0.5**
Org Al	**0.6**	0.2	-0.4	**0.8**	-	-	-	-	-	-	-	-	-	-	1	0.4	**0.5**	**0.7**
Inorg Al	0.3	-0.1	**-0.6**	0.5	-	-	-	-	-	-	-	-	-	-	-	1	0.07	0.1
MeHg	0.3	0.6	0.2	0.09	-	-	-	-	-	-	-	-	-	-	-	-	1	**0.9**
THg	**0.6**	0.4	0.05	0.5	-	-	-	-	-	-	-	-	-	-	-	-	-	1

Table 13: Changes in fish THg concentrations in four common species between 2000s and 2010s at 43 lakes in New York State. Regional averages include standard error, and n_{20XX} columns indicate the number of samples used from each period. Percent change in bold indicates a $p \leq 0.05$.

Region	Lake	Yellow perch			Walleye			Smallmouth bass			Largemouth bass		
		Percent change%	**n_{2000s}**	**n_{2010s}**	**Percent change%**	**n_{2000s}**	**n_{2010s}**	**Percent change%**	**n_{2000s}**	**n_{2010s}**	**Percent change%**	**n_{2000s}**	**n_{2010s}**
Northeast	Big Moose Lake	-7	96	13									
	Delta Lake	**44**	29	40	**-17**	9	37	2	11	41			
	Fall Lake	**-30**	31	23									
	Ferris Lake	**48**	45	25									
	Francis Lake	-12	71	20									
	Hinckley Reservoir	29	10	7				**30**	10	14			
	Kings Flow	5	45	16									
	Limekiln Lake	-8	67	37									

Long Lake	37	10	28									
Middle Stoner Lake							**-34**	10	8			
North Lake	**170**	60	44									
Payne Lake										**108**	6	11
Red Lake	-6	10	10	**-35**	2	6	**-38**	8	10			
Rock Pond	**37**	75	20									
Sand Lake	**-53**	59	9									
Soft Maple Dam Pond	-14	29	2									
Spy Lake							**77**	10	11			
Sunday Lake	**-33**	71	14									
West Caroga Lake	**25**	44	29				**-23**	15	6			

	Willis Lake	16	9	5									
	Regional mean	15 ± 12			-26 ± 9			2 ±18			108		
Southeast	Canadarago Lake	**118**	20	8	-6	20	10	**-56**	10	2	**53**	15	3
	East Sidney Reservoir	**-28**	9	9				**-49**	5	3			
	Forge Pond	**45**	4	9							-20	20	10
	Fort Pond	**-54**	11	20	**-32**	10	12	**-16**	10	10			
	Fresh Pond										**-20**	10	10
	Goodyear Lake	**190**	10	14	16	8	4				**75**	21	5
	Greenwood Lake	4	10	12	**-75**	10	7						
	Lake Huntington										**-20**	6	3
	Lake Superior	**-80**	10	20									
	Mongaup Pond							-6	10	3			

	North-south Lake	**-50**	10	13									
	Onteora Lake	**-74**	10	7							**-76**	7	7
	Otsego Lake	**-52**	11	11	**86**	10	9	**-59**	8	16			
	Rio Reservoir	-28	10	10							**-73**	2	23
	Swinging Bridge Reservoir	6	9	4	9	10	11	**-56**	9	10			
	Walton Pond	9	10	10									
	White Pond	**-86**	8	5	28	3	9						
	Regional mean	-3 ± 23			4 ± 19			-40 ± 10			-11 ± 22		
West	Cuba Lake	**38**	10	10	**45**	10	10	**-61**	10	10			
	Eaton Brook Reservoir										-12	10	3
	Lake Moraine	**77**	10	6									

Rushford Lake	-2	8	10	**24**	20	20				
Seneca Lake	36	10	10							
Silver Lake	27	10	11	4	10	10		**-30**	10	10
Regional mean	35 ± 12			24 ± 12			-61	-21 ± 9		

10. Appendix 1

	Comparison Criteria	Average % Recovery	Standard deviation
Blanks	<0.2	BDL	NA
CCV	80-120%	95	9.71
MDL	80-120%	102	10.95
OPR	80-120%	108	8

Table A 1: QA/QC percent recoveries from THg in water analysis including blanks, continuous calibration verification (CCV), minimum detection limit (MDL) and ongoing precision recovery (OPR)

	Comparison Criteria	Average	Standard Deviation
Sample RPD/RSD	<15	13.0	9.7
MS RPD/RSD	<20	7.1	5.9
MS %Recovery	67-149%	97.5	15.1

Table A 2: Relative percent difference and relative standard difference for duplicated/triplicated samples and matrix spikes. Also, matrix spike percent recoveries for THg in water analysis

	Comparison Criteria	Average % Recovery	Standard deviation
Blanks	<0.02	BDL	NA
CCV	80-120%	98	20
MDL	80-120%	94	8
OPR	80-120%	100	7

Table A 3: QA/QC percent recoveries from MeHg in water analysis including blanks, continuous calibration verification (CCV), minimum detection limit (MDL) and ongoing precision recovery (OPR)

	Comparison Criteria	Average	Standard Deviation
Sample RPD/RSD	<15	9	4.0
MS RPD/RSD	<20	14.7	5.8
MS %Recovery	67-149%	106	16.7

Table A 4: Relative percent difference and relative standard difference for duplicated/triplicated samples and matrix spikes. Also, matrix spike percent recoveries for MeHg in water analysis.

11. Appendix 2

	Comparison Criteria	Average	SD
CCB	<0.2	BDL	NA
CCV	80-120%	102	9.1
mdl	80-120%	103	13.5
OPR	80-120%	103	10.7
MS	80-120%	106	15.8
Lobster	80-120%	101	10.1
Mussel	80-120%	102	11.7

Table A 5: QA/QC results for analysis of MeHg in invertebrate digests. Showing results for continuous calibration blanks (CCB), continues calibration verification (CCV), minimum detection limit (MDL), ongoing precision recovery (OPR) and matrix spike (MS) samples and analysis of digested standard reference samples (lobster and mussel tissue).

12. Appendix 3

	Comparison Criteria	Average	SD
Blanks	<0.3	0.03	0.05
CCV	80-120%	102	4.1
QCS	80-120%	96	8.8
MS	80-120%	101	34.1
Duplicate	<20%	3.49	3.43
Triplicate	<35%	3.65	2.45

Table A 6: QA/QC results for analysis of fish including blanks, continuous calibration verification (CCV), quality control sample (QCS), matrix spike (MS) sample duplicates and sample triplicates.

13. References

Aberg, B., L. Ekman, R. Falk, U. Greitz, G. Persson, and J. Snihs (**1969**). Metabolism of methyl mercury (203 Hg) compounds in man. *Archives of Environmental Health: An International Journal* 19:478–484. doi: 10.1080/00039896.1969.10666872

Aiken, G. R., D. M. McKnight, R. L. Wershaw, and P. MacCarthy (**1985**). Humic Substances in Soil, Sediment, and Water. In Humic Substances in Soil, Sediment, and Water. Wiley. doi: 10.1097/00010694-198611000-00011

Åkerblom, S., M. Nilsson, J. Yu, B. Ranneby, and K. Johansson (**2012**). Temporal change estimation of mercury concentrations in northern pike (Esox lucius L.) in Swedish lakes. *Chemosphere* 86:439–445. doi: 10.1016/j.chemosphere.2011.09.037

Anderson, C., and G. Cabana (**2007**). Estimating the trophic position of aquatic consumers in river food webs using stable nitrogen isotopes. *Journal of the North American Benthological Society* 26:273–285. doi: 10.1899/0887-3593(2007)26[273:ETTPOA]2.0.CO;2

Andersson, S., I. Nilsson, and I. Valeur (**1999**). Influence of dolomitic lime on DOC and DON leaching in

a forest soil. *Biogeochemistry* 47:297–317. doi: 10.1007/BF00992911

Appelberg, M., and E. Degerman (**1991**). Development and Stability of Fish Assemblages after Lime Treatment. *Canadian Journal of Fisheries and Aquatic Sciences* 48:546–554. doi: 10.1139/f91-069

Appling, A. P., M. C. Leon, and W. H. McDowell (**2015**). Reducing bias and quantifying uncertainty in watershed flux estimates: the R package loadflex. *Ecosphere* 6:269. doi: http://dx.doi.org/10.1890/ES14-00517.1

Baker, R. F., P. J. Blanchfield, M. J. Paterson, R. J. Flett, and L. Wesson (**2004**). Evaluation of Nonlethal Methods for the Analysis of Mercury in Fish Tissue. *Transactions of the American Fisheries Society* 133:568–576. doi: 10.1577/T03-012.1

Battles, J. J., T. J. Fahey, C. T. Driscoll, J. D. Blum, and C. E. Johnson (**2014**). Restoring Soil Calcium Reverses Forest Decline. *Environmental Science and Technology Letters* 1:15–19. doi: 10.1021/ez400033d

Benoit, J. M. M., C. C. C. Gilmour, A. Heyes, R. P. Mason, and C. L. Miller (**2002**). Geochemical and biological controls over methylmercury production and degradation in aquatic ecosystems. In ACS symposium. pp. 262–297. doi: 10.1021/bk-2003-0835.ch019

Blackwell, B. D., and C. T. Driscoll (**2015**). Using foliar and forest floor mercury concentrations to assess spatial patterns of mercury deposition. *Environmental Pollution* 202:126–134. doi: 10.1016/j.envpol.2015.02.036

Bodaly, R. A., J. W. M. Rudd, R. J. P. Fudge, and C. A. Kelly (**1993**). Mercury Concentrations in Fish Related to Size of Remote Canadian Shield Lakes. *Canadian Journal of Fisheries and Aquatic Sciences* 50:980–987. doi: 10.1139/f93-113

Bradley, D. C., and S. J. Ormerod (**2002**). Long-term effects of catchment liming on invertebrates in upland streams. *Freshwater Biology* 47:161–171. doi: 10.1046/j.1365-2427.2002.00770.x

Bravo, A. G., S. Bouchet, J. Tolu, E. Björn, A. Mateos-Rivera, and S. Bertilsson (**2017**). Molecular composition of organic matter controls methylmercury formation in boreal lakes. *Nature Communications* 8:14255. doi: 10.1038/ncomms14255

Burgess, N. M., and M. W. Meyer (**2008**). Methylmercury exposure associated with reduced productivity in common loons. *Ecotoxicology* 17:83–91. doi: 10.1007/s10646-007-0167-8

Burns, D. A., M. R. McHale, C. T. Driscoll, and K. M. Roy (**2006**). Response of surface water chemistry to reduced levels of acid precipitation: Comparison of trends in two regions of New York, USA. *Hydrological Processes* 20:1611–1627. doi: 10.1002/hyp.5961

Burns, D. A., K. Riva-Murray, P. M. Bradley, G. R. Aiken, and M. E. Brigham (**2012**). Landscape controls on total and methyl Hg in the upper Hudson River basin, New York, USA. *Journal of Geophysical Research: Biogeosciences* 117:n/a-n/a. doi: 10.1029/2011JG001812

Chen, C. Y., and C. L. Folt (**2005a**). High plankton densities reduce mercury biomagnification. *Environmental Science and Technology* 39:115–121. doi: 10.1021/es0403007

Chen, C. Y., and C. L. Folt (**2005b**). High plankton densities reduce mercury biomagnification. *Environmental Science and Technology* 39:115–121. doi: 10.1021/es0403007

Chen, H., R. C. Johnston, B. F. Mann, R. K. Chu, N. Tolic, J. M. Parks, and B. Gu (**2017**). Identification

of mercury and dissolved organic matter complexes using ultrahigh resolution mass spectrometry. *Environmental Science & Technology Letters* 4:59–65. doi: 10.1021/acs.estlett.6b00460

Chen, J., B. Gu, E. J. LeBoeuf, H. Pan, and S. Dai (**2002**). Spectroscopic characterization of the structural and functional properties of natural organic matter fractions. *Chemosphere* 48:59–68. doi: 10.1016/S0045-6535(02)00041-3

Chen, L., and C. T. Driscoll (**2005**). Regional assessment of the response of the acid−base status of lake watersheds in the adirondack region of New York to changes in atmospheric deposition using PnET-BGC. *Environmental Science & Technology* 39:787–794. doi: 10.1021/es049583t

Cho, Y., C. T. Driscoll, and J. D. Blum (**2009**). The effects of a whole-watershed calcium addition on the chemistry of stream storm events at the Hubbard Brook Experimental Forest in NH, USA. *Science of The Total Environment* 407:5392–5401. doi: 10.1016/j.scitotenv.2009.06.030

Cirmo, C. P., and C. T. Driscoll (**1996**). The impacts of a watershed CaCO3 treatment on stream and wetland biogeochemistry in the Adirondack Mountains. *Biogeochemistry* 32:265–297. doi: 10.1007/BF02187142

Clair, T. A., and A. Hindar (**2005**). Liming for the mitigation of acid rain effects in freshwaters: A review of recent results. *Environmental Reviews* 13:91–128. doi: 10.1139/a05-009

Clair, T. A., and A. Hindar (**2015**). Liming for the mitigation of acid rain effects in freshwaters: A review of recent results. *Dissertation Materials*.

Clark, J. M., S. H. Bottrell, C. D. Evans, D. T. Monteith, R. Bartlett, R. Rose, R. J. Newton, and P. J. Chapman (**2010**). The importance of the relationship between scale and process in understanding long-term DOC dynamics. *Science of The Total Environment* 408:2768–2775. doi: 10.1016/j.scitotenv.2010.02.046

Clayton, J. L., E. S. Dannaway, R. Menendez, H. W. Rauch, J. J. Renton, S. M. Sherlock, and P. E. Zurbuch (**1998**). Application of Limestone to Restore Fish Communities in Acidified Streams. *North American Journal of Fisheries Management* 18:347–360. doi: 10.1577/1548-8675(1998)018<0347:AOLTRF>2.0.CO;2

Climate Science Special Report (**2017**). Climate Science Special Report: Fourth National Climate Assessment, Volume I. [Online.] Available at https://science2017.globalchange.gov/. doi: 10.7930/J0J964J6

Coleman Wasik, J. K., D. R. Engstrom, C. P. J. Mitchell, E. B. Swain, B. A. Monson, S. J. Balogh, J. D. Jeremiason, B. A. Branfireun, R. K. Kolka, and J. E. Almendinger (**2015**). The effects of hydrologic fluctuation and sulfate regeneration on mercury cycling in an experimental peatland. *Journal of Geophysical Research: Biogeosciences* 120:1697–1715. doi: 10.1002/2015JG002993

Cory, R. M., and D. M. McKnight (**2005**). Fluorescence Spectroscopy Reveals Ubiquitous Presence of Oxidized and Reduced Quinones in Dissolved Organic Matter. *Environmental Science & Technology* 39:8142–8149. doi: 10.1021/es0506962

Dalziel, T. R. K., E. J. Wilson, and M. V. Proctor (**1994**). The effectiveness of catchment liming in restoring acid waters at Loch Fleet, Galloway, Scotland. *Forest Ecology and Management* 68:107–117. doi: 10.1016/0378-1127(94)90142-2

Dennis, I. F., T. A. Clair, C. T. Driscoll, N. Kamman, A. Chalmers, J. Shanley, S. A. Norton, and S. Kahl

(**2005**). Distribution Patterns of Mercury in Lakes and Rivers of Northeastern North America. *Ecotoxicology* 14:113–123. doi: 10.1007/s10646-004-6263-0

DeWild, J. F., M. L. Olson, and S. D. Olund (**2002**). Determination of methyl mercury by aqueous phase ethylation, followed by gas chromatographic separation with cold vapor atomic fluorescence detection. *U.S Geological Survey, Open-File Report.*

Dickinson, N. R. (**1983**). A division of southern and western New York State into ecological zones.

Dittman, J. A. (**2010**). Mercury dynamicsin streams, lakes, and fish in the northeastern, United States. *Dissertation Materials*.

Dittman, J. A., and C. T. Driscoll (**2009**). Factors influencing changes in mercury concentrations in lake water and yellow perch (Perca flavescens) in Adirondack lakes. *Biogeochemistry* 93:179–196. doi: 10.1007/s10533-009-9289-9

Dittman, J. A., J. B. Shanley, C. T. Driscoll, G. R. Aiken, A. T. Chalmers, J. E. Towse, and P. Selvendiran (**2010**). Mercury dynamics in relation to dissolved organic carbon concentration and quality during high flow events in three northeastern U.S. streams. *Water Resources Research* 46:W07522. doi: 10.1029/2009WR008351

Dittmar, T., B. Koch, N. Hertkorn, and G. Kattner (**2008**). A simple and efficient method for the solid-phase extraction of dissolved organic matter (SPE-DOM) from seawater. *Limnology and Oceanography: Methods* 6:230–235. doi: 10.4319/lom.2008.6.230

Dong, W., Y. Bian, L. Liang, and B. Gu (**2011**). Binding Constants of Mercury and Dissolved Organic Matter Determined by a Modified Ion Exchange Technique. *Environmental Science & Technology* 45:3576–3583. doi: 10.1021/es104207g

Drevnick, P. E., D. E. Canfield, P. R. Gorski, A. L. C. Shinneman, D. R. Engstrom, D. C. G. Muir, G. R. Smith, P. J. Garrison, L. B. Cleckner, J. P. Hurley, R. B. Noble, et al. (**2007**). Deposition and cycling of sulfur controls mercury accumulation in Isle Royale fish. *Environmental Science and Technology* 41:7266–7272. doi: 10.1021/es0712322

Drevnick, P. E., D. R. Engstrom, C. T. Driscoll, E. B. Swain, S. J. Balogh, N. C. Kamman, D. T. Long, D. G. C. Muir, M. J. Parsons, K. R. Rolfhus, and R. Rossmann (**2012**). Spatial and temporal patterns of mercury accumulation in lacustrine sediments across the Laurentian Great Lakes region. *Environmental Pollution* 161:252–260. doi: 10.1016/j.envpol.2011.05.025

Driscoll, C. T., C. P. Cirmo, T. J. Fahey, V. L. Blette, P. A. Bukaveckas, D. A. Burns, C. P. Gubala, D. J. Leopold, R. M. Newton, D. J. Raynal, C. L. Schofield, et al. (**1996**). The experimental watershed liming study: Comparison of lake and watershed neutralization strategies. *Biogeochemistry* 32:143–174. doi: 10.1007/BF02187137

Driscoll, C. T., K. M. Driscoll, H. Fakhraei, and K. Civerolo (**2016**). Long-term temporal trends and spatial patterns in the acid-base chemistry of lakes in the Adirondack region of New York in response to decreases in acidic deposition. *Atmospheric Environment* 146:5–14. doi: 10.1016/j.atmosenv.2016.08.034

Driscoll, C. T., K. M. Driscoll, M. J. Mitchell, and D. J. Raynal (**2003**). Effects of acidic deposition on forest and aquatic ecosystems in New York State. *Environmental Pollution* 123:327–336. doi: 10.1016/S0269-7491(03)00019-8

Driscoll, C. T., K. M. Driscoll, K. M. Roy, and J. Dukett (**2007a**). Changes in the chemistry of lakes in the Adirondack region of New York following declines in acidic deposition. *Applied Geochemistry* 22:1181–1188. doi: 10.1016/j.apgeochem.2007.03.009

Driscoll, C. T., C. Y. Han, Y.Chen, D. C. Evers, K. F. Lambert, T. M. Holsen, N. C. Kamman, and R. K. Munson (**2007b**). Mercury Contamination in Forest and Freshwater Ecosystems in the Northeastern United States. *BioScience* 57:17. doi: 10.1641/B570106

Driscoll, C. T., G. B. Lawrence, A. J. Bulger, T. J. Butler, C. S. Cronan, C. Eagar, K. F. Lambert, G. E. Likens, J. L. Stoddard, and K. C. Weathers (**2001**). Acidic deposition in the northeastern United States: sources and inputs, ecosystem effects, and management strategies. *BioScience* 51:180.

Driscoll, C. T., R. P. Mason, H. M. Chan, D. J. Jacob, and N. Pirrone (**2013**). Mercury as a global pollutant: Sources, pathways, and effects. *Environmental Science and Technology* 47:4967–4983. doi: 10.1021/es305071v

Driscoll, C. T., C. Yan, C. L. Schofield, R. K. Munson, and J. Holsapple (**1994**). The Mercury cycle and fish in the Adirondacks. *Environmental science & technology* 28:136–143.

ESRI (**2017**). ArcGIS Desktop.

Essington, T. E., and J. N. Houser (**2003**). The Effect of Whole-Lake Nutrient Enrichment on Mercury Concentration in Age-1 Yellow Perch. *Transactions of the American Fisheries Society* 132:57–68. doi: 10.1577/1548-8659(2003)132<0057:TEOWLN>2.0.CO;2

Evans, C. D., P. J. Chapman, J. M. Clark, D. T. Monteith, and M. S. Cresser (**2006**). Alternative explanations for rising dissolved organic carbon export from organic soils. *Global Change Biology* 12:2044–2053. doi: 10.1111/j.1365-2486.2006.01241.x

Fakhraei, H., C. T. Driscoll, P. Selvendiran, J. V. DePinto, J. Bloomfield, S. Quinn, and H. C. Rowell (**2014**). Development of a total maximum daily load (TMDL) for acid-impaired lakes in the Adirondack region of New York. *Atmospheric Environment* 95:277–287. doi: 10.1016/j.atmosenv.2014.06.039

Fiorentino, I., T. J. Fahey, P. M. Groffman, C. T. Driscoll, C. Eagar, and T. G. Siccama (**2003**). Initial responses of phosphorus biogeochemistry to calcium addition in a northern hardwood forest ecosystem. *Canadian Journal of Forest Research* 33:1864–1873. doi: 10.1139/x03-111

Fransman, B., and B. Nihlgaard (**1995**). Water chemistry in forested catchments after topsoil treatment with liming agents in South Sweden. *Water, Air, & Soil Pollution* 85:895–900. doi: 10.1007/BF00476943

French, T. D., A. J. Houben, J.-P. W. Desforges, L. E. Kimpe, S. V Kokelj, A. J. Poulain, J. P. Smol, X. Wang, and J. M. Blais (**2014**). Dissolved organic carbon thresholds affect mercury bioaccumulation in Arctic lakes. *Environmental Science and Technology* 48:3162–3168. doi: 10.1021/es403849d

Fuss, C. B., C. T. Driscoll, and J. L. Campbell (**2015**). Recovery from chronic and snowmelt acidification: Long-term trends in stream and soil water chemistry at the Hubbard Brook Experimental Forest, New Hampshire, USA. *Journal of Geophysical Research: Biogeosciences* 120:2360–2374. doi: 10.1002/2015JG003063

George, S. D., B. P. Baldigo, G. B. Lawrence, and R. L. Fuller (**2018**). Effects of watershed and in-stream liming on macroinvertebrate communities in acidified tributaries to an Adirondack lake. *Ecological*

Indicators 85:1058–1067. doi: 10.1016/j.ecolind.2017.11.048

Gerson, J. R., C. T. Driscoll, J. D. Demers, A. K. Sauer, B. D. Blackwell, M. R. Montesdeoca, J. B. Shanley, and D. S. Ross (**2017**). Deposition of mercury in forests across a montane elevation gradient: Elevational and seasonal patterns in methylmercury inputs and production. *Journal of Geophysical Research: Biogeosciences* 122:1922–1939. doi: 10.1002/2016JG003721

Gerson, J. R., C. T. Driscoll, and K. M. Roy (**2016**). Patterns of nutrient dynamics in Adirondack lakes recovering from acid deposition. *Ecological Applications* 26:1758–1770. doi: 10.1890/15-1361.1

Giang, A., and N. E. Selin (**2016**). Benefits of mercury controls for the United States. *Proceedings of the National Academy of Sciences* 113:286–291. doi: 10.1073/pnas.1514395113

Gilmour, C. C., G. S. Riedel, M. C. Ederington, J. T. Bell, J. M. Benoit, G. A. Gill, and M. C. Stordal (**1998**). Methylmercury concentrations and production rates across a trophic gradient in the northern Everglades. *Biogeochemistry* 40:327–345. doi: 10.1023/A:1005972708616

Gomez-Saez, G. V., J. Niggemann, T. Dittmar, A. M. Pohlabeln, S. Q. Lang, A. Noowong, T. Pichler, L. Wörmer, and S. I. Bühring (**2016**). Molecular evidence for abiotic sulfurization of dissolved organic matter in marine shallow hydrothermal systems. *Geochimica et Cosmochimica Acta* 190:35–52. doi: 10.1016/j.gca.2016.06.027

Gorski, P. R., D. E. Armstrong, J. P. Hurley, and D. P. Krabbenhoft (**2008**). Influence of natural dissolved organic carbon on the bioavailability of mercury to a freshwater alga. *Environmental Pollution* 154:116–123. doi: 10.1016/j.envpol.2007.12.004

Graham, A. M., G. R. Aiken, and C. C. Gilmour (**2012**). Dissolved organic matter enhances microbial mercury methylation under sulfidic conditions. *Environmental Science and Technology* 46:2715–2723. doi: 10.1021/es203658f

Graham, A. M., G. R. Aiken, and C. C. Gilmour (**2013**). Effect of Dissolved Organic Matter Source and Character on Microbial Hg Methylation in Hg−S−DOM Solutions. doi: 10.1021/es400414a

Grandjean, P., and M. Bellanger (**2017**). Calculation of the disease burden associated with environmental chemical exposures: application of toxicological information in health economic estimation. *Environmental Health* 16:123. doi: 10.1186/s12940-017-0340-3

Grieve, I. C. (**1990**). Effects of catchment liming and afforestation on the concentration and fractional composition of aluminium in the Loch Fleet catchment, SW Scotland. *Journal of Hydrology* 115:385–396. doi: 10.1016/0022-1694(90)90216-K

Haitzer, M., G. R. Aiken, and J. N. Ryan (**2002**). Binding of mercury(II) to dissolved organic matter: The role of the mercury-to-DOM concentration ratio. *Environmental Science and Technology* 36:3564–3570. doi: 10.1021/es025699i

Haitzer, M., G. R. Aiken, and J. N. Ryan (**2003**). Binding of mercury(II) to aquatic humic substances: Influence of pH and source of humic substances. *Environmental Science and Technology* 37:2436–2441. doi: 10.1021/es026291o

Håkanson, L., Å. Nilsson, and T. Andersson (**1988**). Mercury in fish in Swedish lakes. *Environmental Pollution* 49:145–162. doi: 10.1016/0269-7491(88)90246-1

Halfman, J. D. (**2017**). Decade-Scale Water Quality Variability in the Eastern Finger Lakes , New York. *Clearwaters*.

Hall, L. W., W. D. Killen, and S. A. Fischer (**1994**). The efficacy of a limestone doser to mitigate stream acidification in a Maryland coastal plain stream: Implication for migratory fish species. *Environmental Monitoring and Assessment* 31:233–257. doi: 10.1007/BF00577256

Hayhoe, K., C. P. Wake, T. G. Huntington, L. Luo, M. D. Schwartz, J. Sheffield, E. Wood, B. Anderson, J. Bradbury, A. DeGaetano, T. J. Troy, and D. Wolfe (**2007a**). Past and future changes in climate and hydrological indicators in the US Northeast. *Climate Dynamics* 28:381–407. doi: 10.1007/s00382-006-0187-8

Hayhoe, K., C. P. Wake, T. G. Huntington, L. Luo, M. D. Schwartz, J. Sheffield, E. Wood, B. Anderson, J. Bradbury, A. DeGaetano, T. J. Troy, and D. Wolfe (**2007b**). Past and future changes in climate and hydrological indicators in the US Northeast. *Climate Dynamics* 28:381–407. doi: 10.1007/s00382-006-0187-8

Herrero Ortega, S., N. Catalán, E. Björn, H. Gröntoft, T. G. Hilmarsson, S. Bertilsson, P. Wu, K. Bishop, O. Levanoni, and A. G. Bravo (**2017**). High methylmercury formation in ponds fueled by fresh humic and algal derived organic matter. *Limnology & Oceanography*. doi: 10.1002/lno.10722

Hertkorn, N., M. Frommberger, M. Witt, B. P. Koch, P. Schmitt-Kopplin, and E. M. Perdue (**2008**). Natural organic matter and the event horizon of mass spectrometry. *Analytical Chemistry* 80:8908–8919. doi: 10.1021/ac800464g

Hill, W. R., and I. L. Larsen (**2005**). Growth Dilution of Metals in Microalgal Biofilms. *Environmental Science & Technology* 39:1513–1518. doi: 10.1021/es049587y

Hindar, A., R. F. Wright, P. Nilsen, T. Larssen, and R. Høgberget (**2003**). Effects on stream water chemistry and forest vitality after whole-catchment application of dolomite to a forest ecosystem in southern Norway. *Forest Ecology and Management* 180:509–525. doi: 10.1016/S0378-1127(02)00647-3

Hintelmann, H., R. Harris, A. Heyes, J. P. Hurley, C. A. Kelly, D. P. Krabbenhoft, S. Lindberg, J. W. M. Rudd, K. J. Scott, and V. L. St. Louis (**2002**). Reactivity and mobility of new and old mercury deposition in a boreal forest ecosystem during the first year of the METAALICUS study. *Environmental Science and Technology* 36:5034–5040. doi: 10.1021/es025572t

Hodgkins, G. A., R. W. Dudley, and T. G. Huntington (**2003**). Changes in the timing of high river flows in New England over the 20th Century. *Journal of Hydrology* 278:244–252. doi: 10.1016/S0022-1694(03)00155-0

Hongve, D., S. Haaland, G. Riise, I. Blakar, and S. Norton (**2012**). Decline of acid rain enhances mercury concentrations in fish. *Environmental science & technology* 46:2490–1. doi: 10.1021/es3002629

Hudy, M., D. M. Downey, and D. W. Bowman (**2000**). Successful Restoration of an Acidified Native Brook Trout Stream through Mitigation with Limestone Sand. *North American Journal of Fisheries Management* 20:453–466. doi: 10.1577/1548-8675(2000)020<0453:SROAAN>2.3.CO;2

Huntington, T. G., G. A. Hodgkins, B. D. Keim, and R. W. Dudley (**2004**). Changes in the Proportion of Precipitation Occurring as Snow in New England (1949–2000). *Journal of Climate* 17:2626–2636. doi: 10.1175/1520-0442(2004)017<2626:CITPOP>2.0.CO;2

Jeffries, D. S., T. G. Brydges, P. J. Dillon, and W. Keller (**2003**). Monitoring the Results of Canada/U.S.A. Acid Rain Control Programs: Some Lake Responses. *Environmental Monitoring and Assessment* 88:3–19. doi: 10.1023/A:1025563400336

Jiang, T., A. G. Bravo, U. Skyllberg, E. Björn, D. Wang, H. Yan, and N. W. Green (**2018**). Influence of dissolved organic matter (DOM) characteristics on dissolved mercury (Hg) species composition in sediment porewater of lakes from southwest China. *Water Research* 146:146–158. doi: 10.1016/j.watres.2018.08.054

Johnson, C. E., C. T. Driscoll, J. D. Blum, T. J. Fahey, and J. J. Battles (**2014**). Soil chemical dynamics after calcium silicate addition to a Northern hardwood forest. *Soil Science Society of America Journal* 78:1458. doi: 10.2136/sssaj2014.03.0114

Josephson, D. C., J. M. Robinson, J. Chiotti, K. J. Jirka, and C. E. Kraft (**2014**). Chemical and biological recovery from acid deposition within the Honnedaga Lake watershed, New York, USA. *Environmental Monitoring and Assessment* 186:4391–4409. doi: 10.1007/s10661-014-3706-9

Karimi, R., N. S. Fisher, and C. L. Folt (**2010**). Multielement Stoichiometry in Aquatic Invertebrates: When Growth Dilution Matters. *The American Naturalist* 176. doi: 10.5061/dryad.1858

Kerin, E. J., C. C. Gilmour, E. Roden, M. T. Suzuki, J. D. Coates, and R. P. Mason (**2006**). Mercury methylation by dissimilatory iron-reducing bacteria. *Applied and Environmental Microbiology* 72:7919–7921. doi: 10.1128/AEM.01602-06

Kim, S., R. W. Kramer, and P. G. Hatcher (**2003**). Graphical Method for Analysis of Ultrahigh-Resolution Broadband Mass Spectra of Natural Organic Matter, the Van Krevelen Diagram. *Analytical Chemistry* 75:5336–5344. doi: 10.1021/ac034415p

Knight, A., S. P. Bhavsar, B. A. Branfireun, P. Drouin, R. Prashad, S. Petro, and M. Oke (**2019**). A comparison of fish tissue mercury concentrations from homogenized fillet and nonlethal biopsy plugs. *Journal of Environmental Sciences* 80:137–145. doi: 10.1016/j.jes.2018.12.004

Koch, B. P., T. Dittmar, M. Witt, and G. Kattner (**2007**). Fundamentals of molecular formula assignment to ultrahigh resolution mass data of natural organic matter. *Analytical Chemistry* 79:1758–1763. doi: 10.1021/ac061949s

Koch, B. P., M. Witt, R. Engbrodt, T. Dittmar, and G. Kattner (**2005**). Molecular formulae of marine and terrigenous dissolved organic matter detected by electrospray ionization Fourier transform ion cyclotron resonance mass spectrometry. *Geochimica et Cosmochimica Acta* 69:3299–3308. doi: 10.1016/j.gca.2005.02.027

Kolka, R. K., C. E. Riggs, E. A. Nater, T. R. Wickman, E. L. Witt, and J. T. Butcher (**2019**). Temporal fluctuations in young-of-the-year yellow perch mercury bioaccumulation in lakes of northeastern Minnesota. *Science of the Total Environment* 656:475–481. doi: 10.1016/j.scitotenv.2018.11.280

Ksionzek, K. B., O. J. Lechtenfeld, S. L. McCallister, P. Schmitt-Kopplin, J. K. Geuer, W. Geibert, and B. P. Koch (**2016**). Dissolved organic sulfur in the ocean: Biogeochemistry of a petagram inventory. *Science* 354:456–459. doi: 10.1126/science.aaf7796

Laudon, H., J. Buttle, S. K. Carey, J. McDonnell, K. McGuire, J. Seibert, J. Shanley, C. Soulsby, and D. Tetzlaff (**2012**). Cross-regional prediction of long-term trajectory of stream water DOC response to climate change. *Geophysical Research Letters* 39:4–9. doi: 10.1029/2012GL053033

Lawrence, G. B., D. A. Burns, and K. Riva-murray (**2016**). A new look at liming as an approach to accelerate recovery from acidic deposition effects deposition effects. doi: 10.1016/j.scitotenv.2016.03.176

Lawrence, G. B., K. M. Roy, B. P. Baldigo, H. a Simonin, S. B. Capone, J. W. Sutherland, S. a Nierzwicki-Bauer, and C. W. Boylen (**2008**). Chronic and episodic acidification of Adirondack streams from acid rain in 2003-2005. *Journal of environmental quality* 37:2264–74. doi: 10.2134/jeq2008.0061

Lawrence, G. B., H. A. Simonin, B. P. Baldigo, K. M. Roy, and S. B. Capone (**2011**). Changes in the chemistry of acidified Adirondack streams from the early 1980s to 2008. *Environmental Pollution* 159:2750–2758. doi: 10.1016/j.envpol.2011.05.016

Likens, G. E., and D. C. Buso (**2012**). Dilution and the elusive baseline. *Environmental Science and Technology* 46:4382–4387. doi: 10.1021/es3000189

Mangal, V., B. R. Stenzler, A. J. Poulain, and C. Gue (**2019**). Aerobic and Anaerobic Bacterial Mercury Uptake is Driven by Algal Organic Matter Composition and Molecular Weight´. doi: 10.1021/acs.est.8b04909

Mangal, V., N. L. Stock, and C. GuÃ©guen (**2016**). Molecular characterization of phytoplankton dissolved organic matter (DOM) and sulfur components using high resolution Orbitrap mass spectrometry. *Analytical and Bioanalytical Chemistry* 408:1891–1900. doi: 10.1007/s00216-015-9295-9

Mao, H., Z. Ye, and C. Driscoll (**2017**). Meteorological effects on Hg wet deposition in a forested site in the Adirondack region of New York during 2000–2015. *Atmospheric Environment* 168:90–100. doi: 10.1016/j.atmosenv.2017.08.058

Matthews, D. A., D. B. Babcock, J. G. Nolan, A. R. Prestigiacomo, S. W. Effler, C. T. Driscoll, S. G. Todorova, and K. M. Kuhr (**2013**). Whole-lake nitrate addition for control of methylmercury in mercury-contaminated Onondaga Lake, NY. *Environmental Research* 125:52–60. doi: 10.1016/j.envres.2013.03.011

McAvoy, D. C., R. C. Santore, J. D. Shosa, and C. T. Driscoll (**1992**). Comparison between pyrocatechol violet and 8-hydroxyquinonline procedures for determining aluminum fractions.

McCabe, G. J., and D. M. Wolock (**2002**). A step increase in streamflow in the conterminous United States. *Geophysical Research Letters* 29:38-1-38–4. doi: 10.1029/2002GL015999

Menendez, R., J. L. Clayton, and P. E. Zurbuch (**1996**). Chemical and Fishery Responses to Mitigative Liming of an Acidic Stream, Dogway Fork, West Virginia. *Restoration Ecology* 4:220–233. doi: 10.1111/j.1526-100X.1996.tb00175.x

Millard, G. D., C. Driscoll, D. A. Burns, M. R. Montesdeoca, and K. Riva-Murray (**2018a**). Response of mercury in an Adirondack (NY, USA) forest stream to watershed lime application. *Environmental Science: Processes & Impacts*:8–12. doi: 10.1039/C7EM00520B

Millard, G. D., C. T. Driscoll, D. A. Burns, M. R. Montesdeoca, and K. Riva-Murray (**2018b**). Response of mercury in an Adirondack (NY, USA) forest stream to watershed lime application. *Environmental Science: Processes and Impacts* 20:607–620. doi: 10.1039/c7em00520b

Minimata Convention on Mercury (**2017**). Minimata Convention. [Online.] Available at http://www.mercuryconvention.org/Convention/tabid/3426/language/en-US/Default.aspx.

Mitchell, C. P. J., and C. C. Gilmour (**2008**). Methylmercury production in a Chesapeake Bay salt marsh. *Journal of Geophysical Research* 113:1–14. doi: 10.1029/2008JG000765

Monson, B. A., D. F. Staples, S. P. Bhavsar, T. M. Holsen, C. S. Schrank, S. K. Moses, D. J. McGoldrick, S. M. Backus, and K. A. Williams (**2011**). Spatiotemporal trends of mercury in walleye and largemouth bass from the Laurentian Great Lakes Region. *Ecotoxicology* 20:1555–1567. doi: 10.1007/s10646-011-0715-0

Monteith, D. T., J. L. Stoddard, C. D. Evans, H. a de Wit, M. Forsius, T. Høgåsen, A. Wilander, B. L. Skjelkvåle, D. S. Jeffries, J. Vuorenmaa, B. Keller, et al. (**2007**). Dissolved organic carbon trends resulting from changes in atmospheric deposition chemistry. *Nature* 450:537–540. doi: 10.1038/nature06316

Moreau, J. W., C. M. Gionfriddo, D. P. Krabbenhoft, J. M. Ogorek, J. F. DeWild, G. R. Aiken, and E. E. Roden (**2015**). The effect of natural organic matter on mercury methylation by Desulfobulbus propionicus 1pr3. *Frontiers in Microbiology* 6. doi: 10.3389/fmicb.2015.01389

National Drought Mitigation Center, U.S. Department of Agriculture, and U.S. National Oceanic and Atmospheric Administration (**2016**). U.S. Drought Monitor. *2016*. [Online.] Available at https://droughtmonitor.unl.edu.

Newton, R. M., D. A. Burns, V. L. Blette, and C. T. Driscoll (**1996**). Effect of whole catchment liming on the episodic acidification of two adirondack streams. *Biogeochemistry* 32:299–322. doi: 10.1007/BF02187143

NIDIS (**2019**). Drought in New York. *Drought.gov*. [Online.] Available at https://www.drought.gov/drought/states/new-york.

Nurmi, J. T., and P. G. Tratnyek (**2002**). Electrochemical Properties of Natural Organic Matter (NOM), Fractions of NOM, and Model Biogeochemical Electron Shuttles. *Environmental Science & Technology* 36:617–624. doi: 10.1021/es0110731

NYS DOH (**2018**). Health Advice on Eating Fish you Catch. [Online.] Available at https://www.health.ny.gov/environmental/outdoors/fish/health_advisories/.

Olson, M. L., and J. F. DeWild (**1999**). Techniques for the collection and species-specific analysis of low levels of mercury in water, sediment, and biota. *U.S. Geological Survey Water Resource Investigations Rep.*

Peterson, S. A., J. Van Sickle, R. M. Hughes, J. A. Schacher, and S. F. Echols (**2004**). A Biopsy Procedure for Determining Filet and Predicting Whole-Fish Mercury Concentration. *Archives of Environmental Contamination and Toxicology* 48:99–107. doi: 10.1007/s00244-004-0260-4

Pickhardt, P. C., C. L. Folt, C. Y. Chen, B. Klaue, and J. D. Blum (**2002**). Algal blooms reduce the uptake of toxic methylmercury in freshwater food webs. *Proceedings of the National Academy of Sciences of the United States of America* 99:4419–23. doi: 10.1073/pnas.072531099

Pirrone, N., S. Cinnirella, X. Feng, R. B. Finkelman, H. R. Friedli, J. Leaner, R. Mason, a. B. Mukherjee, G. B. Stracher, D. G. Streets, and K. Telmer (**2010**). Global mercury emissions to the atmosphere from anthropogenic and natural sources. *Atmospheric Chemistry and Physics* 10:5951–5964. doi: 10.5194/acp-10-5951-2010

Pohlabeln, A. M., and T. Dittmar (**2015**). Novel insights into the molecular structure of non-volatile marine dissolved organic sulfur. *Marine Chemistry* 168:86–94. doi: 10.1016/j.marchem.2014.10.018

Post, D. M. (**2002**). The long and short of food-chain length. *Trends in Ecology & Evolution* 17:269–277.

doi: 10.1016/S0169-5347(02)02455-2

Poulin, B. A., J. N. Ryan, and G. R. Aiken (**2014**). Effects of Iron on Optical Properties of Dissolved Organic Matter. *Environmental Science & Technology* 48:10098–10106. doi: 10.1021/es502670r

Prestbo, E. M., and D. A. Gay (**2009**). Wet deposition of mercury in the U.S. and Canada, 1996-2005: Results and analysis of the NADP mercury deposition network (MDN). *Atmospheric Environment* 43:4223–4233. doi: 10.1016/j.atmosenv.2009.05.028

R Core Team (**2018**). R: A language and environment for statistical computing. [Online.] Available at https://www.r-project.org/.

Rask, M., R. I. Jones, M. Järvinen, A. Paloheimo, M. Salonen, J. Syväranta, and M. Verta (**2007**). Changes in fish mercury concentrations over 20 years in an acidified lake subject to experimental liming. *Applied Geochemistry* 22:1229–1240. doi: 10.1016/j.apgeochem.2007.03.015

Rice, G. E., J. K. Hammitt, and J. S. Evans (**2010**). A Probabilistic Characterization of the Health Benefits of Reducing Methyl Mercury Intake in the United States. *Environmental Science & Technology* 44:5216–5224. doi: 10.1021/es903359u

Riva-Murray, K., P. M. Bradley, L. C. Chasar, D. T. Button, M. E. Brigham, B. C. Scudder Eikenberry, C. A. Journey, and M. A. Lutz (**2013**). Influence of dietary carbon on mercury bioaccumulation in streams of the Adirondack Mountains of New York and the Coastal Plain of South Carolina, USA. *Ecotoxicology* 22:60–71. doi: 10.1007/s10646-012-1003-3

Riva-Murray, K., L. C. Chasar, P. M. Bradley, D. A. Burns, M. E. Brigham, M. J. Smith, and T. A. Abrahamsen (**2011**). Spatial patterns of mercury in macroinvertebrates and fishes from streams of two contrasting forested landscapes in the eastern United States. *Ecotoxicology* 20:1530–1542. doi: 10.1007/s10646-011-0719-9

Roy, K. M., J. Duckett, N. Houck, and G. B. Lawrence (**2012**). A Long-Term Monitoring Program for Evaluating Changes in Water Quality in Selected Adirondack Waters. [Online.] Available at http://www.adirondacklakessurvey.org/pubs.shtml.

Runkel, R., and L. De Cicco (**2017**). rloadest: River Load Estimation. R package version 0.4.5.

Sauer, V. B., and D. P. Turnipseed (**2010**). Stage measurement at gaging stations. In U.S. Geological Survey Techniques and Methods 3. p. 45.

Schmidt, K. L., and W. E. Sharpe (**2002**). Passive treatment methods for acid water in Pennsylvania. In. Penn State College of Agricultural Sciences.

Schoch, N., M. J. Glennon, D. C. Evers, M. Duron, A. K. Jackson, C. T. Driscoll, J. W. Ozard, and A. K. Sauer (**2014**). The impact of mercury exposure on the Common Loon (Gavia immer) population in the Adirondack Park, New York, USA. *Waterbirds* 37:133–146. doi: 10.1675/063.037.sp116

Scudder, B. C., L. C. Chasar, L. R. DeWeese, M. E. Brigham, D. A. Wentz, and W. G. Brumbaugh (**2008**). Procedures for Collecting and Processing Streambed Sediment and Pore Water for Analysis of Mercury as Part of the National Water-Quality Assessment Program. *Open-File Report 2008–1279National*:63.

Sebestyen, S. D., E. W. Boyer, and J. B. Shanley (**2009**). Responses of stream nitrate and DOC loadings to hydrological forcing and climate change in an upland forest of the northeastern United States. *Journal of Geophysical Research: Biogeosciences* 114:1–11. doi: 10.1029/2008JG000778

Seigneur, C., and K. Lohman (**2008**). Effect of bromine chemistry on the atmospheric mercury cycle. *Journal of Geophysical Research Atmospheres* 113:1–12. doi: 10.1029/2008JD010262

Selin, N. E. (**2009**). Global biogeochemical cycling of mercury: a review. *Annual Review of Environment and Resources* 34:43–63. doi: 10.1146/annurev.environ.051308.084314

Shao, S., C. T. Driscoll, C. E. Johnson, T. J. Fahey, J. J. Battles, and J. D. Blum (**2016**). Long-term responses in soil solution and stream-water chemistry at Hubbard Brook after experimental addition of wollastonite. *Environmental Chemistry* 13:528. doi: 10.1071/EN15113

Shastri, Y., and U. Diwekar (**2008**). Optimal control of lake pH for mercury bioaccumulation control. *Ecological Modelling* 216:1–17. doi: 10.1016/j.ecolmodel.2008.03.019

Shaw, J. B., T.-Y. Lin, F. E. Leach, A. V Tolmachev, N. Tolić, E. W. Robinson, D. W. Koppenaal, and L. Paša-Tolić (**2016**). 21 Tesla Fourier Transform Ion Cyclotron Resonance Mass Spectrometer Greatly Expands Mass Spectrometry Toolbox. *Journal of The American Society for Mass Spectrometry* 27:1929–1936. doi: 10.1007/s13361-016-1507-9

Simonin, H. A., J. J. Loukmas, L. C. Skinner, and K. M. Roy (**2008a**). Lake variability: Key factors controlling mercury concentrations in New York State fish. *Environmental Pollution* 154:107–115. doi: 10.1016/j.envpol.2007.12.032

Simonin, H., J. Loukmas, L. Skinner, and K. Roy (**2008b**). Strategic Monitoring of Mercury in New York State Fish. [Online.] Available at http://www.dec.ny.gov/docs/wildlife_pdf/hgfish.pdf.

Skyllberg, U. (**2008**). Competition among thiols and inorganic sulfides and polysulfides for Hg and MeHg in wetland soils and sediments under suboxic conditions: Illumination of controversies and implications for MeHg net production. *Journal of Geophysical Research: Biogeosciences* 113:n/a-n/a. doi: 10.1029/2008JG000745

Skyllberg, U., P. R. Bloom, J. Qian, C.-M. Lin, and W. F. Bleam (**2006**). Complexation of Mercury(II) in Soil Organic Matter: EXAFS Evidence for Linear Two-Coordination with Reduced Sulfur Groups. *Environmental Science & Technology* 40:4174–4180. doi: 10.1021/es0600577

Skyllberg, U., J. Qian, W. Frech, K. Xia, and W. F. Bleam (**2003**). Distribution of mercury, methyl mercury and organic sulphur species in soil, soil solution and stream of a boreal forest catchment. *Biogeochemistry* 64:53–76. doi: 10.1023/A:1024904502633

Skyllberg, U., K. Xia, P. R. Bloom, E. A. Nater, and W. F. Bleam (**2000**). Binding of Mercury(II) to Reduced Sulfur in Soil Organic Matter along Upland-Peat Soil Transects. *Journal of Environment Quality* 29:855. doi: 10.2134/jeq2000.00472425002900030022x

Sleighter, R. L., Y. P. Chin, W. A. Arnold, P. G. Hatcher, A. J. McCabe, B. C. McAdams, and G. C. Wallace (**2014**). Evidence of Incorporation of Abiotic S and N into Prairie Wetland Dissolved Organic Matter. *Environmental Science and Technology Letters* 1:345–350. doi: 10.1021/ez500229b

Sleighter, R. L., and P. G. Hatcher (**2007**). The application of electrospray ionization coupled to ultrahigh resolution mass spectrometry for the molecular characterization of natural organic matter. *Journal of Mass Spectrometry*. doi: 10.1002/jms.1221

Streets, D. G., H. M. Horowitz, Z. Lu, L. Levin, C. P. Thackray, and E. M. Sunderland (**2018**). Global And Regional Trends In Mercury Emissions And Concentrations, 2010–2015. *Atmospheric Environment* 201:2010–2015. doi: 10.1016/j.atmosenv.2018.12.031

Streets, D. G., H. M. Horowitz, Z. Lu, L. Levin, C. P. Thackray, and E. M. Sunderland (**2019**). Global and regional trends in mercury emissions and concentrations, 2010–2015. *Atmospheric Environment* 201:417–427. doi: 10.1016/j.atmosenv.2018.12.031

Stubbins, A., R. G. M. Spencer, H. Chen, P. G. Hatcher, K. Mopper, P. J. Hernes, V. L. Mwamba, A. M. Mangangu, J. N. Wabakanghanzi, and J. Six (**2010**). Illuminated darkness: Molecular signatures of Congo River dissolved organic matter and its photochemical alteration as revealed by ultrahigh precision mass spectrometry. *Limnology and Oceanography* 55:1467–1477. doi: 10.4319/lo.2010.55.4.1467

Sundseth, K., J. M. Pacyna, E. G. Pacyna, J. Munthe, M. Belhaj, and S. Astrom (**2010**). Economic benefits from decreased mercury emissions: Projections for 2020. *Journal of Cleaner Production* 18:386–394. doi: 10.1016/j.jclepro.2009.10.017

Syversen, T., and P. Kaur (**2012**). The toxicology of mercury and its compounds. *Journal of Trace Elements in Medicine and Biology* 26:215–226. doi: 10.1016/j.jtemb.2012.02.004

Taylor, M. S., C. T. Driscoll, J. M. Lepak, D. C. Josephson, K. J. Jirka, and C. E. Kraft. (**2019**). Temporal trends in fish mercury concentrations in an Adirondack Lake managed with a persistent predator removal program. *Ecotoxicology*.

Tfaily, M. M., R. K. Chu, N. Tolić, K. M. Roscioli, C. R. Anderton, L. Paša-Tolić, E. W. Robinson, and N. J. Hess (**2015**). Advanced solvent based methods for molecular characterization of soil organic matter by high-resolution mass spectrometry. *Analytical Chemistry* 87:5206–5215. doi: 10.1021/acs.analchem.5b00116

Todorova, S., C. T. Driscoll, D. A. Matthews, and W. E. Steven (**2015**). Zooplankton Community Changes Confound the Biodilution Theory of Methylmercury Accumulation in a Recovering Mercury- Contaminated Lake. 8–13. doi: 10.1021/es5044084

Tolić, N., Y. Liu, A. Liyu, Y. Shen, M. M. Tfaily, E. B. Kujawinski, K. Longnecker, L.-J. Kuo, E. W. Robinson, L. Paša-Tolić, and N. J. Hess (**2017**). Formularity: Software for Automated Formula Assignment of Natural and Other Organic Matter from Ultrahigh-Resolution Mass Spectra. *Analytical Chemistry* 89:12659–12665. doi: 10.1021/acs.analchem.7b03318

Tsui, M. T. K., J. C. Finlay, and E. A. Nater (**2008**). Effects of stream water chemistry and tree species on release and methylation of mercury during litter decomposition. *Environmental Science and Technology* 42:8692–8697. doi: 10.1021/es800956q

Turnipseed, D. P., and V. B. Sauer (**2010**). Discharge measurement at gaging stations. In U.S. Geological Survey Techniques and Methods 3. p. 87.

U.S. EPA (**1995**). Method 1669: Sampling Ambient Water for Trace Metals at EPA Water Quality Criteria Levels. *United States Environmental Protection Agency*:1–42.

U.S. EPA (**2002**). Method 1631, revision E: mercury in water by oxidation, purge and trap, and cold vapor atomic fluorescence spectrometry. *United States Environmental Protection Agency*:1–46.

U.S. EPA (**2007a**). Method 1630 Methyl Mercury in Water by Distillation, Aqueous Ethylation, Purge and Trap, and Cold Vapor Atomic Fluorescence Spectrometry. *United States Environmental Protection Agency*:1–55.

U.S. EPA (**2007b**). Method 7473: Mercury in solids and solutions by thermal decomposition,

amalgamation, and atomic absorption spectrophotometry. *United States Environmental Protection Agency*.

U.S. EPA (**2017**). Mercury and Air Toxics Standards: Clean Power Plants. *Mercury and Air Toxics Standards*. [Online.] Available at https://www.epa.gov/mats/cleaner-power-plants#controls.

UN Environment Programme Chemicals and Health Branch (**2019**). UN Environment, 2019. Global Mercury Assessment 2018.

Ussiri, D. a. N., and C. E. Johnson (**2004**). Sorption of organic carbon fractions by spodosol mineral horizons. *Soil Science Society of America Journal* 68:253. doi: 10.2136/sssaj2004.0253

Waller, K., C. Driscoll, J. Lynch, D. Newcomb, and K. Roy (**2012**). Long-term recovery of lakes in the Adirondack region of New York to decreases in acidic deposition. *Atmospheric Environment* 46:56–64. doi: 10.1016/j.atmosenv.2011.10.031

Warby, R. A. F., C. E. Johnson, and C. T. Driscoll (**2005**). Chemical recovery of surface waters across the northeastern United States from reduced inputs of acidic deposition: 1984-2001. *Environmental Science and Technology* 39:6548–6554. doi: 10.1021/es048553n

Ward, D. M., K. H. Nislow, and C. L. Folt (**2010**). Bioaccumulation syndrome: Identifying factors that make some stream food webs prone to elevated mercury bioaccumulation. *Annals of the New York Academy of Sciences* 1195:62–83. doi: 10.1111/j.1749-6632.2010.05456.x

Watson, S. B., C. Miller, G. Arhonditsis, G. L. Boyer, W. Carmichael, M. N. Charlton, R. Confesor, D. C. Depew, T. O. Höök, S. A. Ludsin, G. Matisoff, et al. (**2016**). The re-eutrophication of Lake Erie: Harmful algal blooms and hypoxia. *Harmful Algae* 56:44–66. doi: 10.1016/j.hal.2016.04.010

Weishaar, J., G. Aiken, B. Bergamaschi, M. Fram, R. Fujii, and K. Mopper (**2003**). Evaluation of specific ultra-violet absorbance as an indicator of the chemical content of dissolved organic carbon. *Environmental Science and Technology* 37:4702–4708. doi: 10.1021/es030360x

Weiss-Penzias, P. S., D. A. Gay, M. E. Brigham, M. T. Parsons, M. S. Gustin, and A. ter Schure (**2016**). Trends in mercury wet deposition and mercury air concentrations across the U.S. and Canada. *Science of the Total Environment* 568:546–556. doi: 10.1016/j.scitotenv.2016.01.061

Will, G., R. Stumvoll, R. Gotie, and E. Smith (**1982**). The Ecological Zones of Northern New York. *New York Fish and Game Journal* 29:1–25.

Wu, Z., R. P. Rodgers, and A. G. Marshall (**2004**). Two- and Three-Dimensional van Krevelen Diagrams: A Graphical Analysis Complementary to the Kendrick Mass Plot for Sorting Elemental Compositions of Complex Organic Mixtures Based on Ultrahigh-Resolution Broadband Fourier Transform Ion Cyclotron Resonance. *Analytical Chemistry* 76:2511–2516. doi: 10.1021/ac0355449

Ye, Z., H. Mao, and C. T. Driscoll (**2019**). Primary effects of changes in meteorology vs. anthropogenic emissions on mercury wet deposition: A modeling study. *Atmospheric Environment* 198:215–225. doi: 10.1016/j.atmosenv.2018.10.052

Yu, X., C. T. Driscoll, M. Montesdeoca, D. Evers, M. Duron, K. Williams, N. Schoch, and N. C. Kamman (**2011**). Spatial patterns of mercury in biota of Adirondack, New York lakes. *Ecotoxicology* 20:1543–1554. doi: 10.1007/s10646-011-0717-y

Zhang, Y., D. J. Jacob, H. M. Horowitz, L. Chen, H. M. Amos, D. P. Krabbenhoft, F. Slemr, V. L. St. Louis, and E. M. Sunderland (**2016**). Observed decrease in atmospheric mercury explained by global

decline in anthropogenic emissions. *Proceedings of the National Academy of Sciences* 113:526–531. doi: 10.1073/pnas.1516312113

Zhou, C., M. D. Cohen, B. A. Crimmins, H. Zhou, T. A. Johnson, P. K. Hopke, and T. M. Holsen (**2017**). Mercury Temporal Trends in Top Predator Fish of the Laurentian Great Lakes from 2004 to 2015: Are Concentrations Still Decreasing? *Environmental Science and Technology* 51:7386–7394. doi: 10.1021/acs.est.7b00982

Zhou, H., C. Zhou, P. K. Hopke, and T. M. Holsen (**2018**). Mercury wet deposition and speciated mercury air concentrations at rural and urban sites across New York state: Temporal patterns, sources and scavenging coefficients. *Science of the Total Environment* 637–638:943–953. doi: 10.1016/j.scitotenv.2018.05.047

Geoffrey Millard Ph.D.

Environmental Engineer/Biogeochemist
gmillard@syr.edu; (315) 854-4441

Engineering & Computer Science
Syracuse University

Professional Preparation

Ph.D., Civil Engineering
Syracuse University, Syracuse, New York, 2019

M.S. Environmental Engineering Science, Syracuse University, Syracuse, New York, 2016

C.A.S. Sustainable Enterprise, Syracuse University, Syracuse, New York, 2016

B.S. Chemistry,
St. Lawrence University, Canton, New York, 2010

Professional Experience

2018/2019 Teaching Assistant, Syracuse University 2018/2019

2016-2018 National Science Foundation Research Trainee Fellow, Syracuse University

2017-2021 Executive Committee Member, Northeaster Ecosystem Research Co-operative (NERC)

2013-2016 Research Assistant, Syracuse University

2011-2013 Research Support Specialist, SUNY ESF

2011-2013 Scientific Technician, Syracuse Center of Excellence

2009-2010 Research Fellow, Dept. of Chemistry, St. Lawrence University

Funded Research

2018 Environmental Molecular Science Laboratory, Funded research proposal, Syracuse University

Awards/Honours

2019 Nemerow Memorial Scholarship, Syracuse University

2006 Canadian Merit Scholarship, St. Lawrence University

Publications

Millard, G.; Driscoll, C.; Yang, Y.; Montesdeoca, M.; Taylor, M; Boucher, S.; Richter, W.; Paul, E.; Parker, C; Yokota, K. (in development) Patterns and trends of fish mercury in New York State. Ecotoxicology.

Millard, G.; Riva-Murray, K.; Driscoll, C.; Burns, D.; Montesdeoca, M. (indevelopment) The impact of lime additions on mercury dynamics in stream chemistry and macroinvertebrates: A comparison of management strategies. Ecotoxicology.

Montesdeoca, M.; Driscoll, C.; **Millard, G.**; Shaw, A. (in development) A comparison of skin on and skin off tissue mercury analysis for different fish species. Ecotoxicology.

Driscoll, C.; Yang, Y.; Gerson, J.; **Millard, G.**; Boucher, S. (in development) Patterns and trends in water and fish mercury in the Adirondacks. Ecotoxicology.

Lautz, L.; McCay, D.; Driscoll, C.; Glas, R.; Gutchess, K.; **Millard, G.**; Johnson, A. (2018) Designing student-centered STEM graduate programs for a multitude of career pathways. EOS. DOI: 10.1029/2018EO101599.

Geoffrey Millard Ph.D.

Environmental Engineer/Biogeochemist
gmillard@syr.edu; (315) 854-4441

Engineering & Computer Science
Syracuse University

Millard, G.; Driscoll, C.; Montesdeoca, M.; Riva-Murray, K.; Burns, D. (2018) Response of mercury in an Adirondack, USA forest stream to watershed lime application: Accelerated recovery from acidification. Environmental Science and Technology. Environmental Science: Processes and Impacts. DOI: 10.1039/C7EM00520B

Buckley, S.; Mitchell, M.; McHale, P.; **Millard, G.** (2014) Variations in carbon dioxide fluxes within a city landscape: Identifying a vehicular influence. Urban Ecosystems. DOI:10.1007/s11252-013-0341-0

Presentations

"Molecular Characterization of Dissolved Organic Matter Components Facilitating the Transport and Transformation of Mercury in Headwater Streams of the Adirondack State Park, New York, USA." **International Conference on Mercury as a Global Pollutant**. Krakow, Poland. September 2019

"Leveraging Established Research Sites for Developing a Multinational Field Methods Course." Poster. **American Geophysical Union**. Washington, DC. December 2018.

"An Introduction to Engineering." Presentation. **Frontiers of Science**. Syracuse, New York. November 2018

"Mercury in the fish and water of unique Lake Kivu, Rwanda." Presentation. **Civil and Environmental Engineering Seminar**. Syracuse, NY. September 2018.

"Monitoring Mercury in New York State Fish." **Syracuse Center of Excellence Symposium**. Presentation. Syracuse, NY. October 2017.

"Response of streamwater mercury concentrations to watershed and in-stream lime applications in an Adirondack, USA watershed." Presentation. **International Conference on Mercury as a Global Pollutant**. Providence, RI. July 2017.

"Response of mercury in a forest stream to lime application: accelerated watershed recovery." Presentation. **Invited Lecture at Brockville Collegiate Institute**. Brockville Ontario Canada. May 2016

"Response of the mercury cycle in an Adirondack USA lake watershed to recovery from decreasing acid deposition and lime application." Presentation. **Ecological Society of America**, Baltimore, MD. Aug 2015.

Synergistic Activities

Advisor, Louis Stokes Alliance for Minority Participation (LSAMP) research program; NASA Scholars Program; Donofrio Scholars Program; Environmental Science and Engineering Research Internships for Undergraduate Students; Environmental Science and Engineering Research Internships for High School Students

Peer mentor, National Science Foundation Education Model Program On Water-Energy Research

Organizer, 2016 Environmental Science and Engineering research discussion group

Geoffrey Millard Ph.D.

Environmental Engineer/Biogeochemist
gmillard@syr.edu; (315) 854-4441

Engineering & Computer Science
Syracuse University

Advisees

Undergraduate Honors Student

1. Michael Persson (2016-2017), Methylmercury and Isotopic Analysis of Invertebrates from a Lime Treated Tributary of Lake Honnedaga

Undergraduate Summer Interns

1. Marcus Bowens (LSAMP; 2014), Predicted effect of lime treatment to soil at Honnedaga Lake
2. Lisa Uwizeyimana (2014, 2015), Improvement on a Correction Factor to Approximate Methyl Mercury Using Specific Ultraviolet Absorvance – SUVA; The Mobility of Mercury and Dissolved organic matter in response to acid, base and calcium sulfate treatments to forest mineral soil
3. Yaskira Mota (2015), Study the effect on Bioavailable Organic Carbon (PBOC) in soil after liming near the Honnedaga Lake in New York State.
4. Yan Chun Chen (Donofrio; 2015), Ultraviolet (UV) Digestion Study for Mercury Analysis in Water
5. Katherine Roskoff (2016), Deposition associated with throughfall along Whiteface Mountain, NY, USA
6. William Guida (2016), The effects of liming on mercury concentration in invertebrates and salamanders on the Honnedaga Lake watershed
7. Kevin Ordoñez (2018), Calcium-based limings and their impact on soil composition and mercury concentration
8. Jessenia Guzmán (NASA; 2018), Evaluating soil quality after a lime application at Honnedaga Lake

High school Summer Interns

1. Francesca Giardine (2016), Phosphorous in Honnedaga Lake, an Acidic Lake in the Adirondacks
2. Lucy Langenberg (2017), Impacts of watershed liming on mercury in Adirondack crayfish
3. Ben Ashby (2018), Mercury in fish and water from the African rift: Lake Kivu, Rwanda

Teaching and Outreach

Achieving data management, visualization and analysis,
Workshop at the 2019 International Conference on Mercury as a Global Pollutant

Applied Microbiology,
Teaching assistant for graduate and undergraduate students. Responsible for grading and laboratory exercises

Introduction to Environmental Engineering,
Teaching assistant for undergraduate course responsible for recitations and grading.

Field Methods Course, Lake Kivu, Kibuye and Gisenyi Rwanda
Developed and assessed water quality activities as part of a multidisciplinary, multinational, field course.

Harmful Algal Blooms in the Finger Lakes, Syracuse, New York
Produced a podcast and website exploring the known science behind the increasing incidence of harmful algal blooms in western New York

Frontiers of Science: Introduction to Engineering, Syracuse New York
Developed and presented active-learning materials for high-achieving, local high school students interested in post-secondary education in STEM field